

# Supporting Information

## DNA Assembly of Modular Components into a Rotatory Nanodevice

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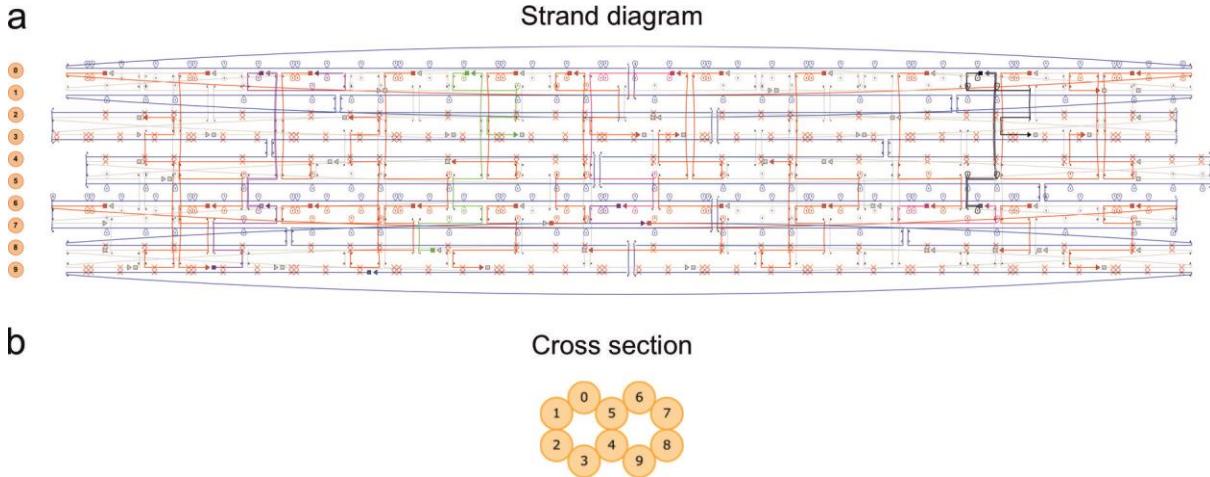
**IV. References**

## I. DNA origami design

**Small origami ring.** The small ring structure was designed using the caDNAno software.<sup>1</sup> It consists of a 10-helix bundle arranged in a ‘honeycomb’ lattice. Constant bending of the DNA bundle was achieved by the deletion and addition of base pairs along the helices. The DNA bundle was split into four parts. Helices of the innermost part (helices 3 and 9, 1<sup>st</sup> layer) have 44 deletions, helices of the 2<sup>nd</sup> layer (helices 2, 4 and 8) have 22 deletions, helices of the 3<sup>rd</sup> layer (helices 1, 5 and 7) have 22 insertions and helices of the 4<sup>th</sup> layer (helices 0 and 6) have 44 insertions. To fold the structure, the p7249 scaffold was used. Due to the small size of the origami, only 2309 bases were used. To prevent the remaining scaffold to disturb the rotation process, the unused part of the scaffold strand points towards the center of the ring. At the end of the DNA bundle, scaffold and staple strand overhangs connect each head and tail region to ensure a closed ring geometry (for more details see Supplementary Figure S1).

**Large origami ring.** The large ring structure was designed with caDNAno<sup>1</sup> software using the p8064 scaffold. The large ring consists of a 12-helix bundle arranged in a ‘honeycomb’ lattice. Constant bending of the DNA bundles was achieved by the deletion and insertion of base pairs along the helices. The DNA bundle was split into six parts. Helices of the innermost part (helices 0 and 11, 1<sup>st</sup> layer) have 90 deletions, helices of the 2<sup>nd</sup> (helices 1 and 10) and 3<sup>rd</sup> part (helices 2 and 9) have 54 and 18 deletions, respectively, helices of the 4<sup>th</sup> (helices 3 and 8), 5<sup>th</sup> (helices 4 and 7) and 6<sup>th</sup> layer (helices 5 and 6) have 18, 54 and 90 insertions, accordingly. At the end of the DNA bundle, staple strand overhangs connect each head and tail region to ensure a closed ring geometry (for more details see Supplementary Figure S2).

## II. Supplementary Figures

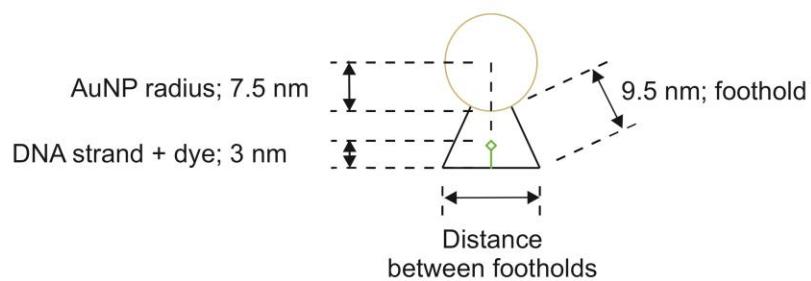


**Supplementary Figure S1.** (a) caDNAno design and strand routing for the small DNA origami ring. Blue: p7249 scaffold; light gray: core staples; red: footholds; green: ATTO550 staples; black: ATTO647N staples; purple: locking strands; pink: foothold strands with locking region at the 3' end. According to whether only the small DNA origami ring or the rotary device was assembled, addition of ATTO647 and locks (black and pink staples) had to be adjusted. (b) Cross section of the DNA bundle layout.



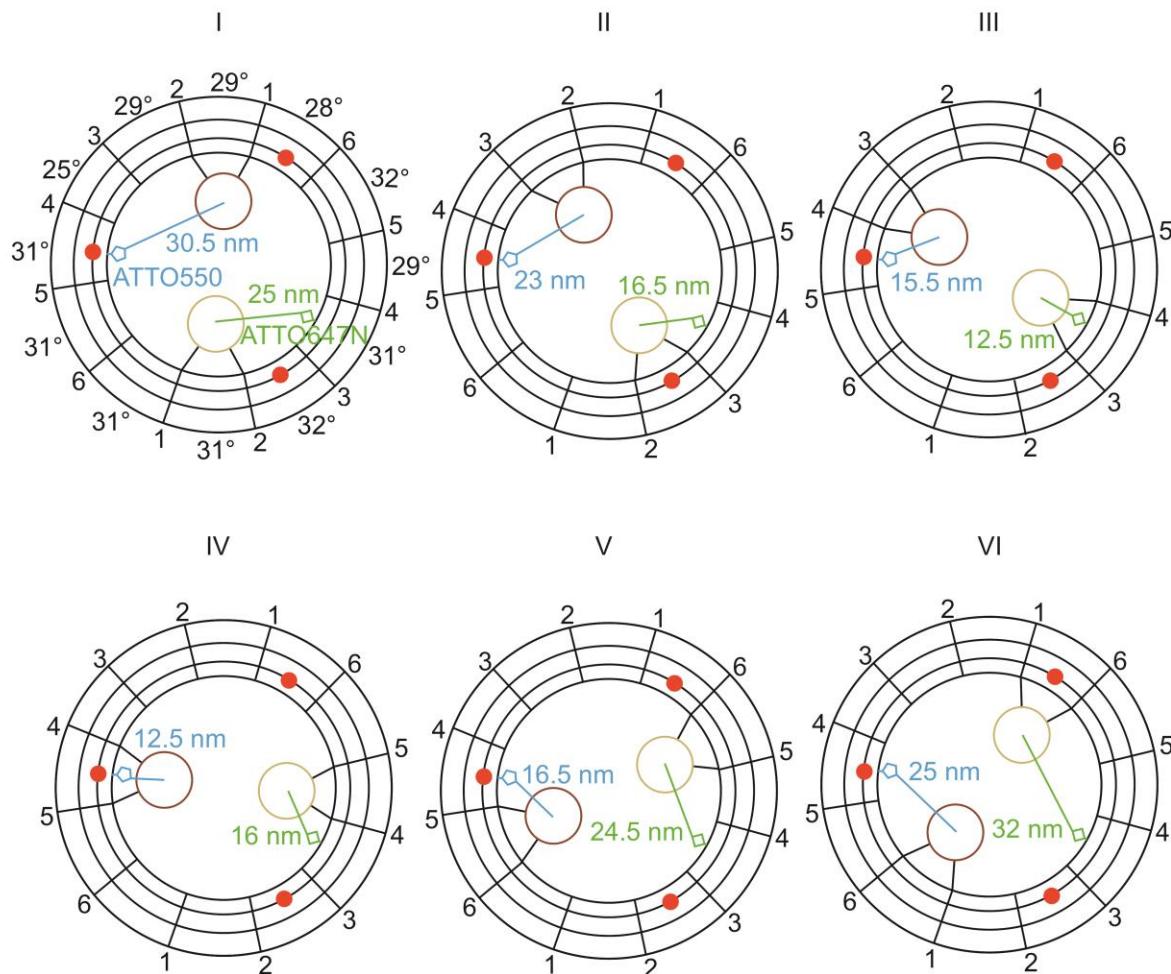
**Supplementary Figure S2.** (a) caDNAno design and strand routing for the large DNA origami ring. Blue: p8064 scaffold; light gray: core staples; red: footholds; green: ATTO647N staples; black: ATTO550 position and locking strand, respectively; purple: locking strands; pink: foothold strands with locking region or ATTO550 hybridization region at the 3' end. According to whether only the large DNA origami ring or the rotary device was assembled, addition of ATTO550 staples and locks (black and pink staples) had to be adjusted. (b) Cross section of the DNA bundle layout.

a

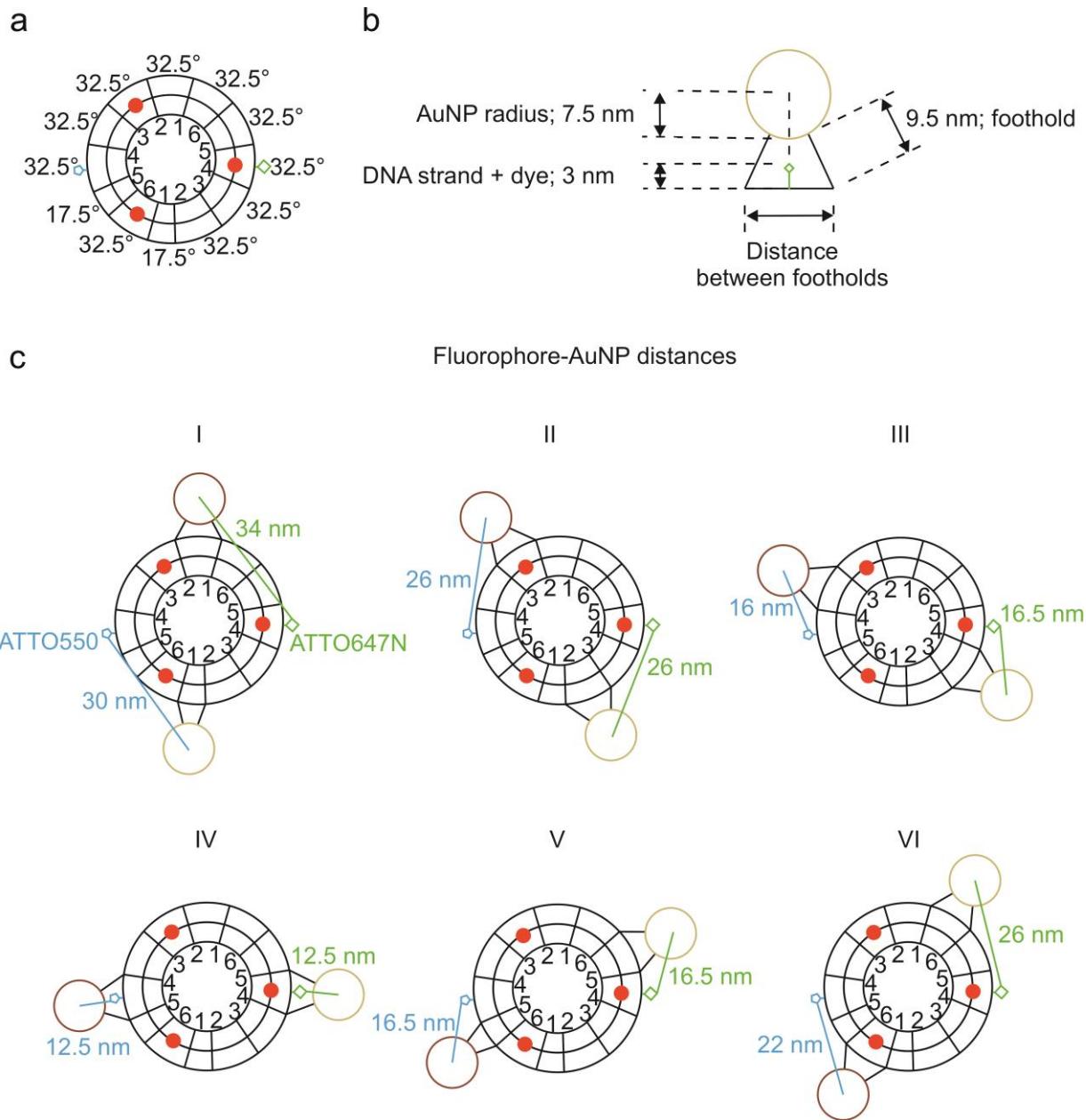


b

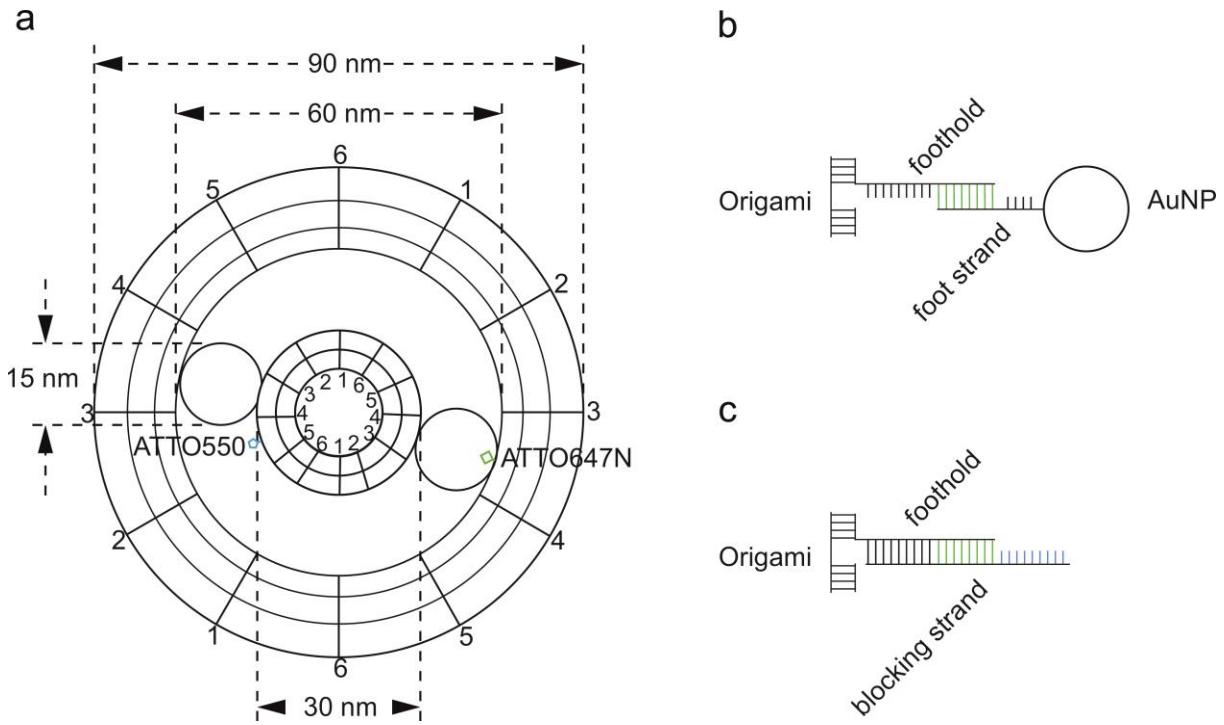
Fluorophore-AuNP distances



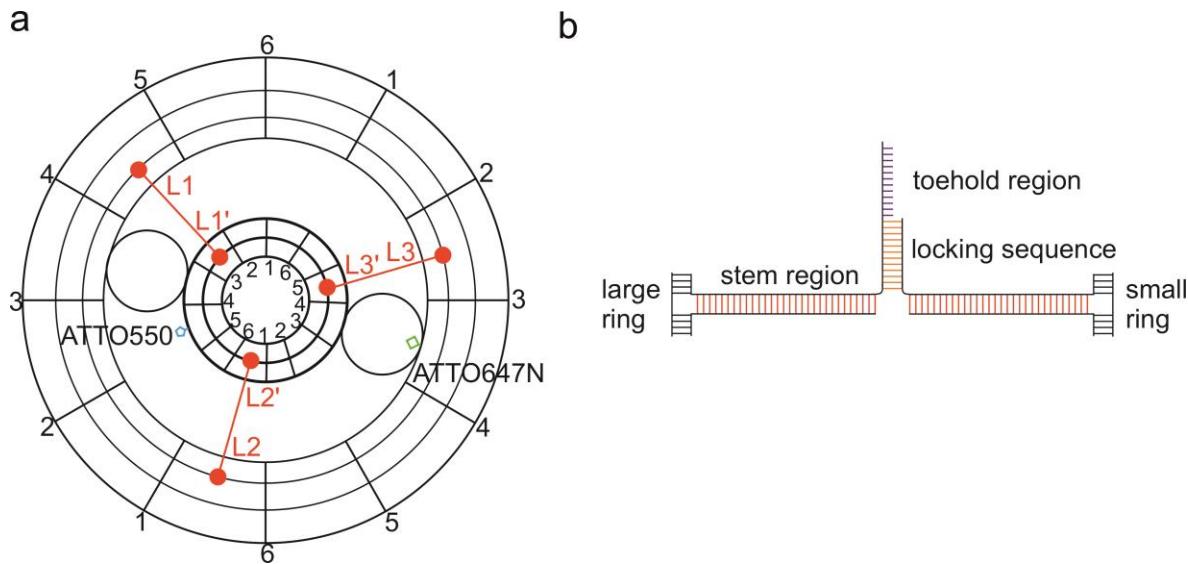
**Supplementary Figure S3.** (a) Illustration of the basic distances. The red dots represent the three locking strand positions. (b) Angles between the foothold rows on the large origami ring and the fluorophore-AuNP distances at different states. The fluorophore-AuNP distances relevant for the theoretical calculations can be found in Supplementary Table S4.



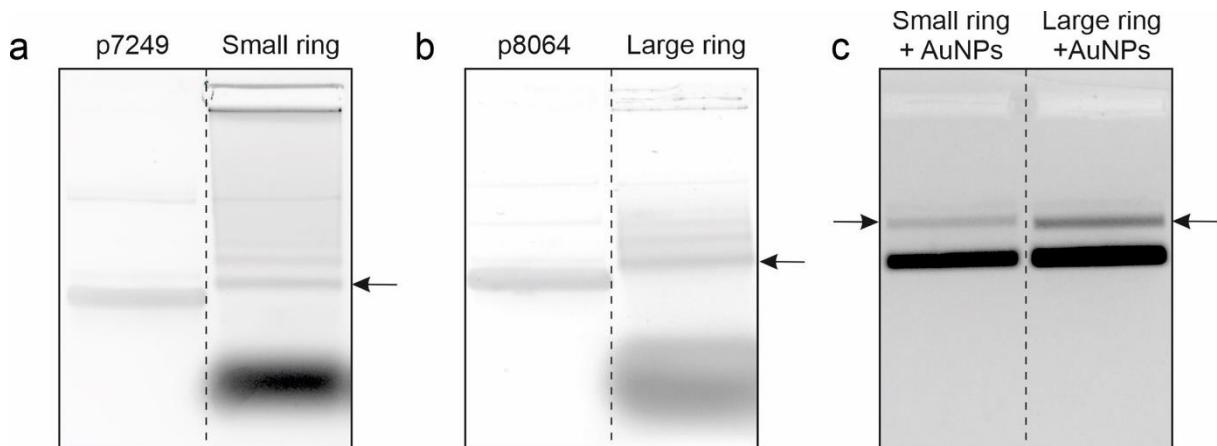
**Supplementary Figure S4.** (a) Angles between the foothold rows on the small origami ring. The positions of the locks are indicated with red dots. (b) Illustration of the basic distances. (c) Fluorophore-AuNP distances at different states. The fluorophore-AuNP distances relevant for the theoretical calculations can be found in Supplementary Table S5.



**Supplementary Figure S5.** (a) Schematic of the planetary gearset nanodevice. The outer and inner diameters of the large ring are ~90 nm and ~60 nm, respectively. The outer diameter of the small ring is ~30 nm. The AuNP diameter is 15 nm. (b) Schematic of the foothold-AuNP interaction. (c) Foothold design and blocking/releasing scheme. Each foothold comprises a unique stem region (black) connected to the DNA origami structure and a AuNP-capture sequence (green) to bind the AuNPs. Blocking strand deactivates the foothold by hybridizing with the stem and AuNP-capturing regions. Each blocking strand further comprises a toehold region (blue). Activation of the foothold is achieved by the addition of the complementary releasing strand, which completely hybridizes with the blocking strand to release it from the foothold.

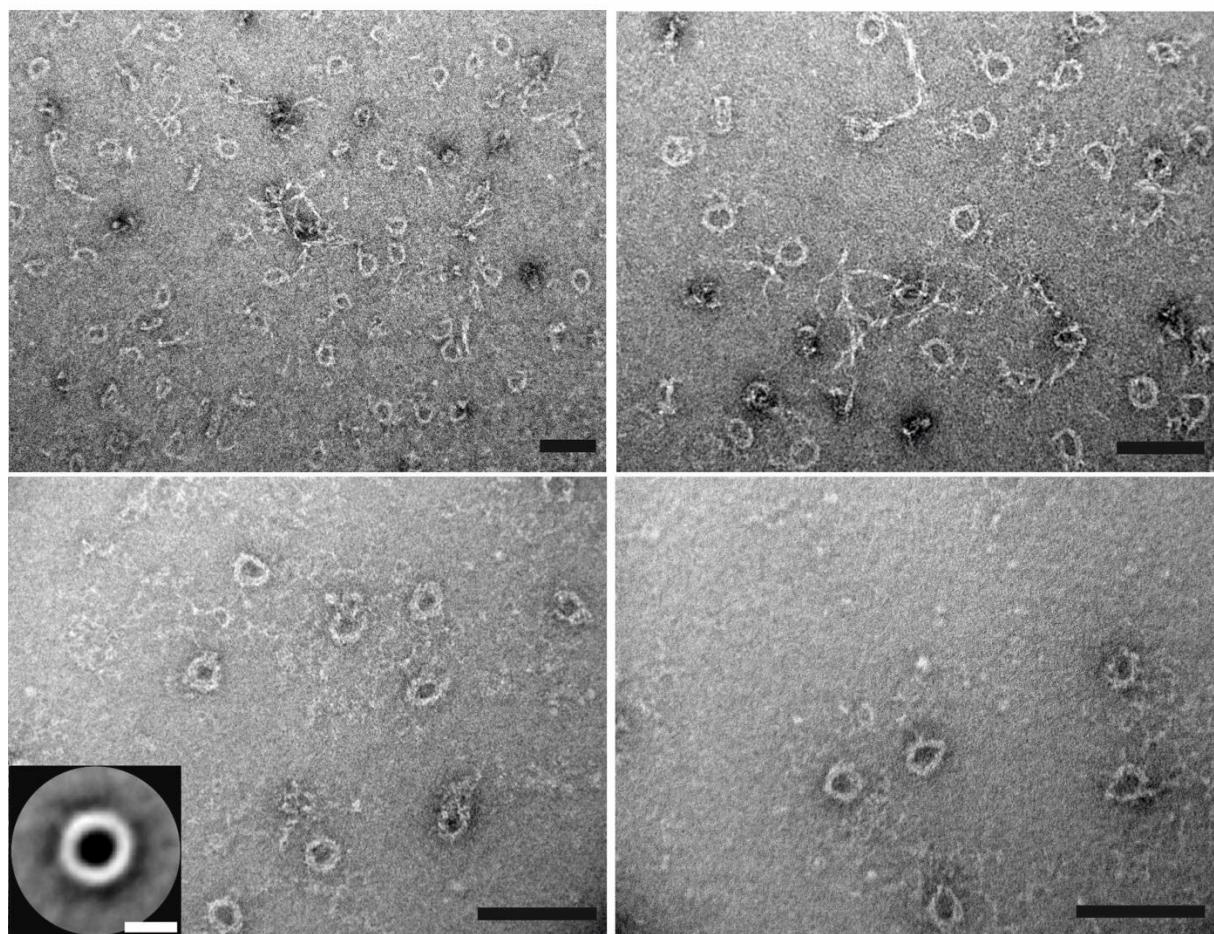


**Supplementary Figure S6.** (a) Schematic of the rotary nanodevice indicated with locking positions. Three orthogonal, complementary locking couples (L1-L1', L2-L2' and L3-L3', red) are placed onto the large and small DNA origami rings to direct the orientation and relative position of the rings. (b) Design of the locking strands. Each lock comprises a stem region (red) and a locking sequence (orange). The linker strand of the large ring further comprises a toehold region (purple) to enable the opening of the locking couple through toehold-mediated strand displacement reactions.

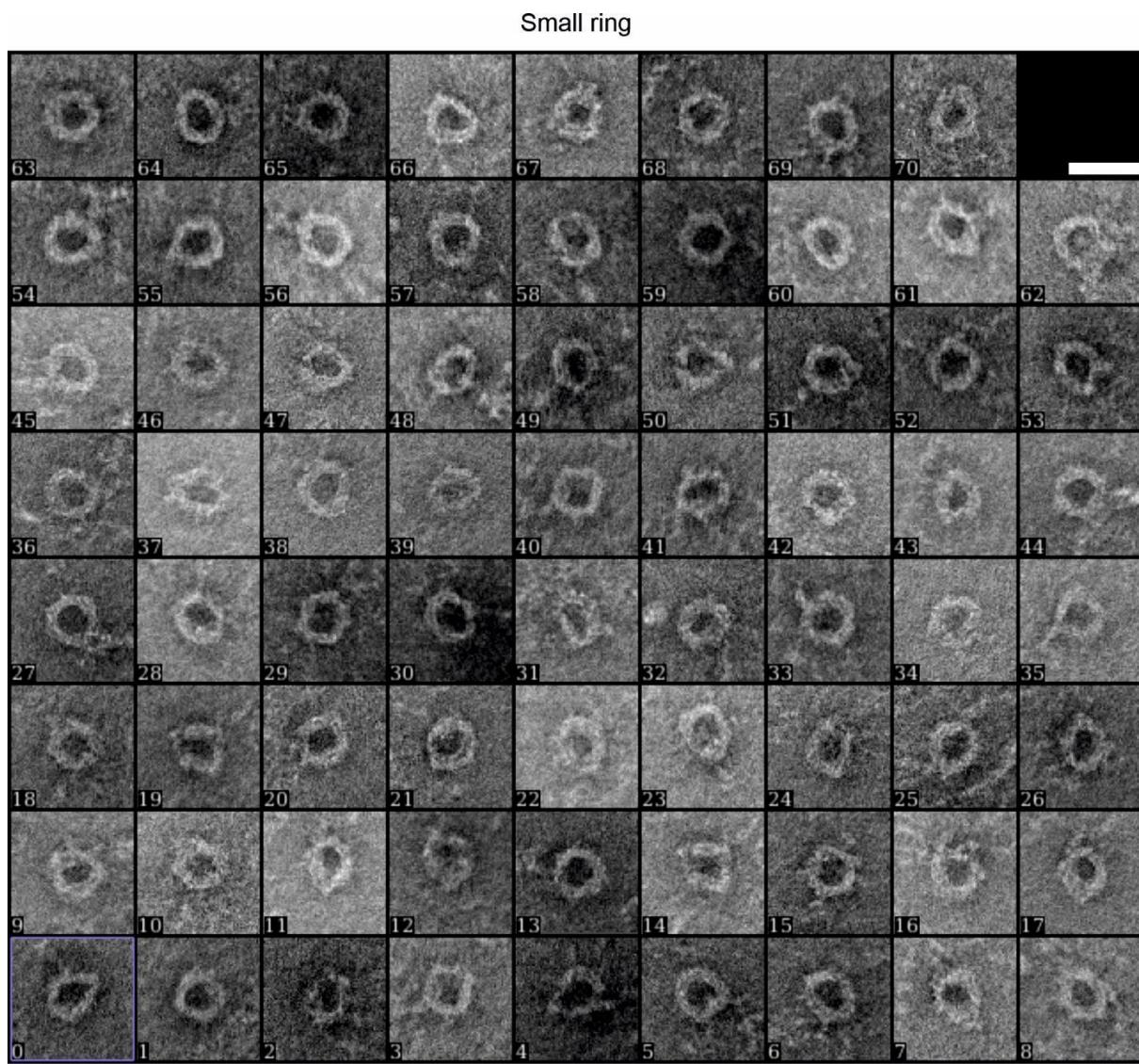


**Supplementary Figure S7.** (a) Agarose gel electrophoresis image of the small DNA origami ring after assembly. The band marked with the arrow contains the desired origami structures. The p7249 scaffold is used as reference. (b) Agarose gel electrophoresis images of the large DNA origami ring after assembly. The band marked with the arrow contains the desired origami structures. The p8064 scaffold is used as reference. (c) Agarose gel electrophoresis image of the AuNP-functionalized small and large origami rings after the annealing process. The bands marked with the arrows contain the desired nanostructures. The intense black bands contain excess AuNPs.

Small ring

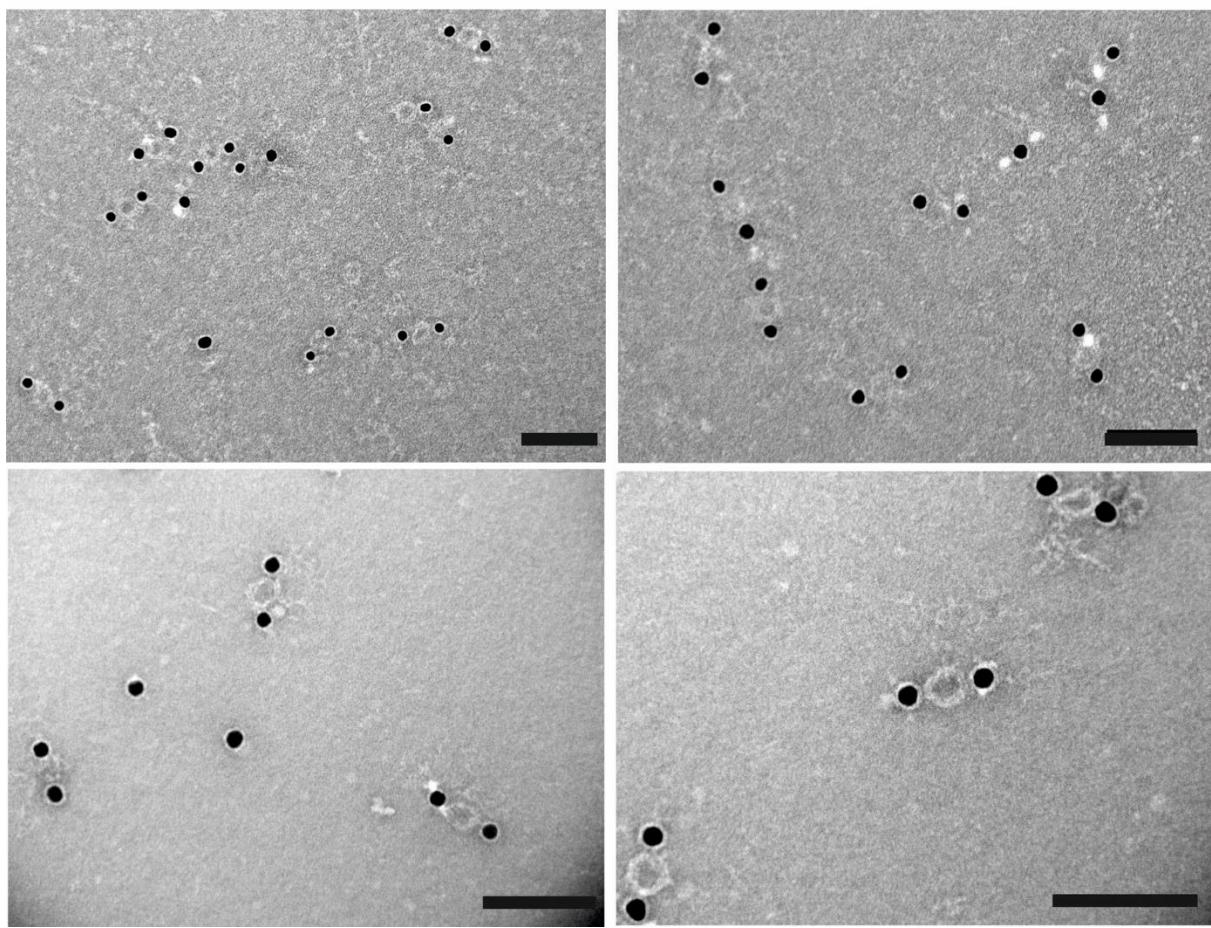


**Supplementary Figure S8.** TEM images of the small origami rings. Scale bar, 100 nm. Inset: averaged TEM image (see Supplementary Figure S9 for the structure library used for the calculation). Scale bar, 20 nm.



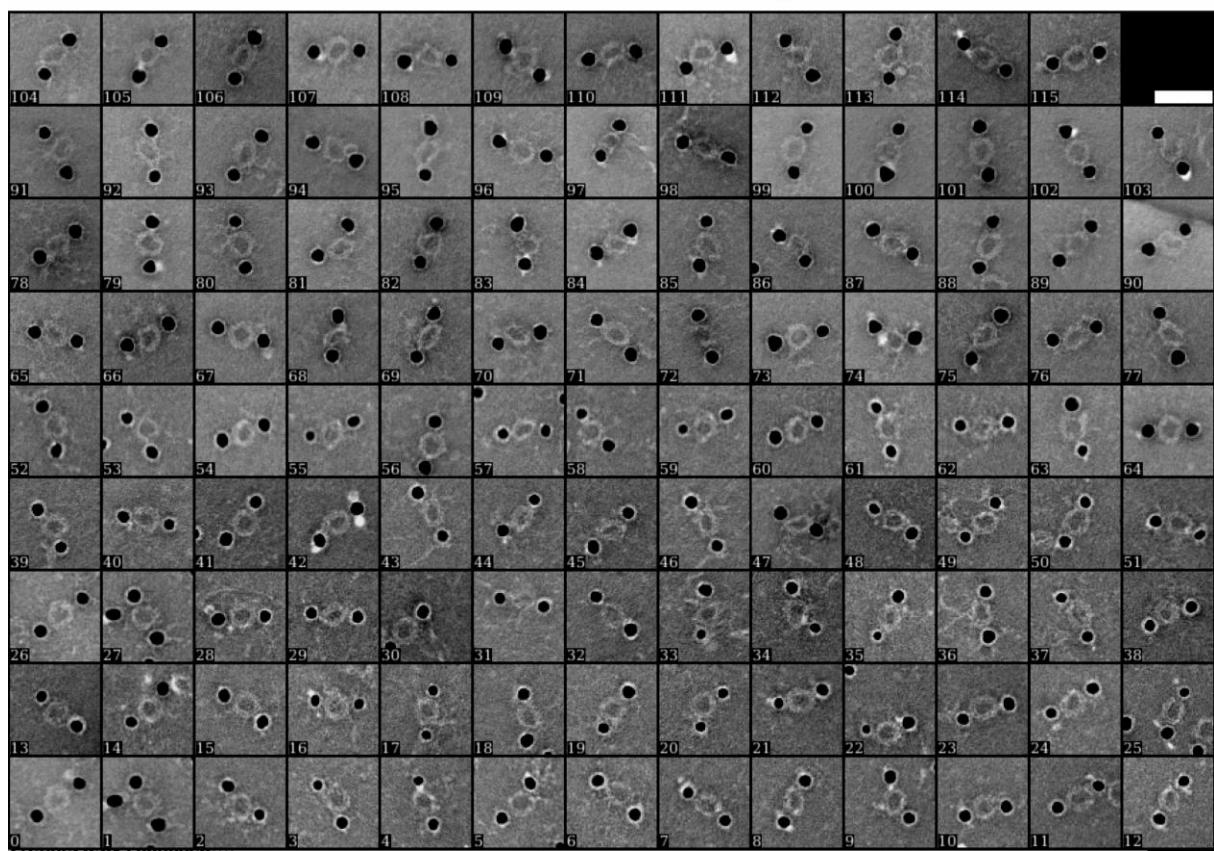
**Supplementary Figure S9.** Structure library of the small origami rings to calculate the averaged TEM image (see Supplementary Figure S8). Scale bar, 50 nm.

AuNP-functionalized small ring



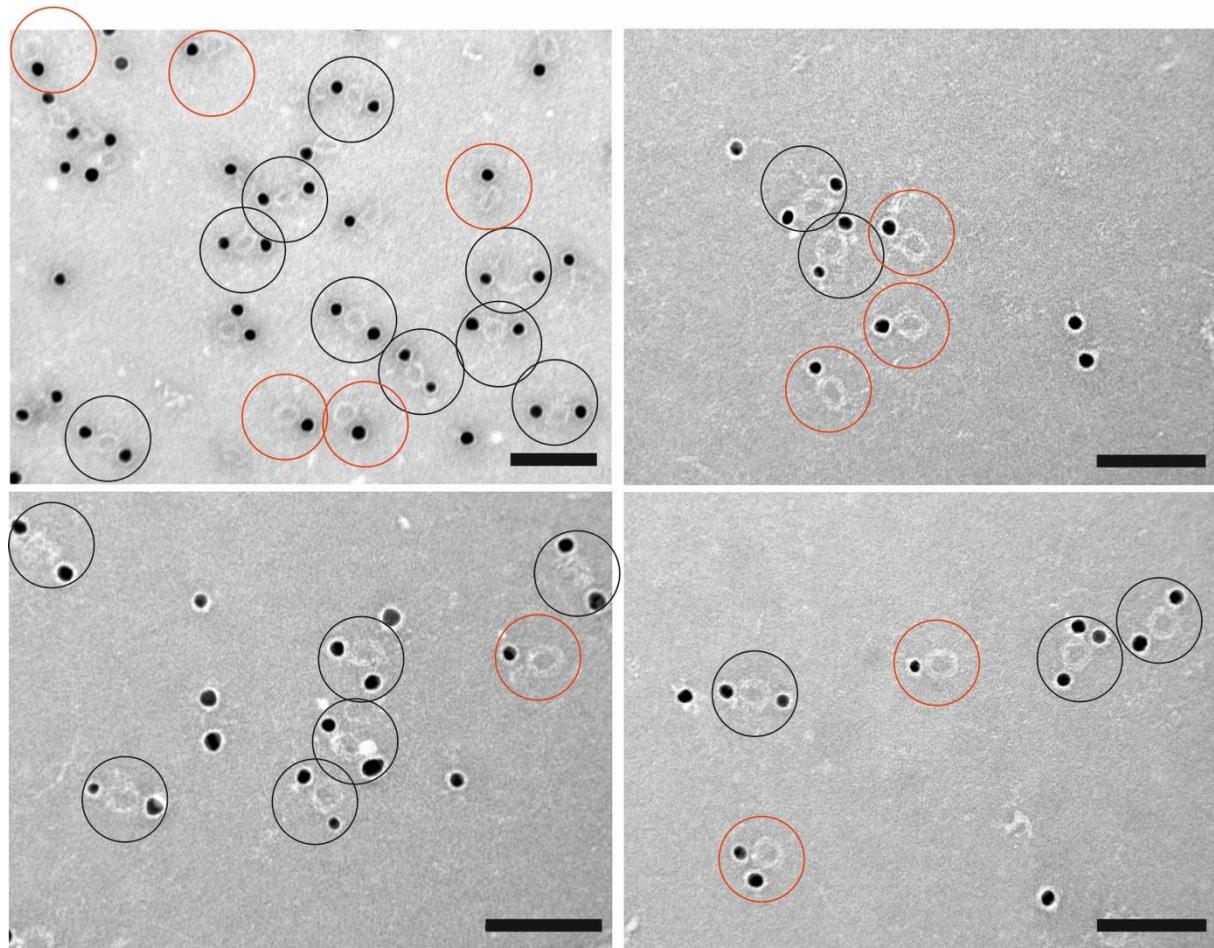
**Supplementary Figure S10.** TEM images of the AuNP-functionalized small origami rings. Scale bar, 100 nm.

AuNP-functionalized small ring

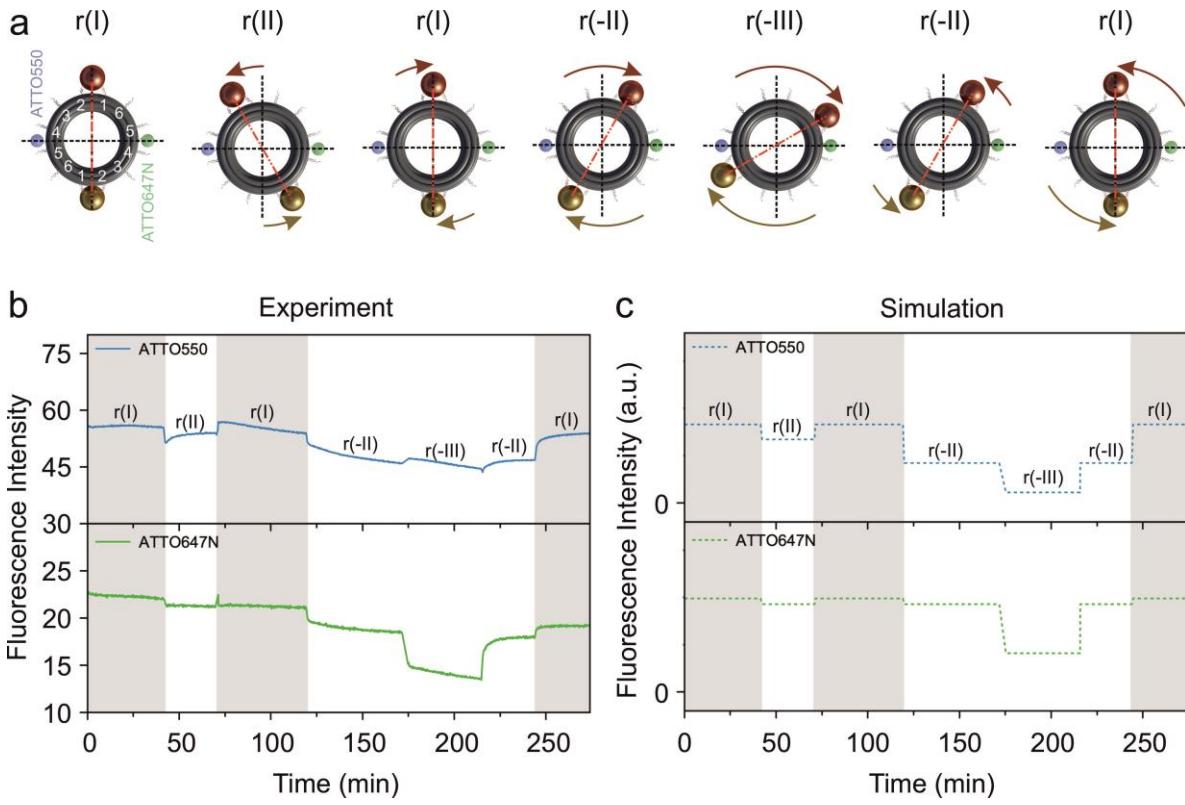


**Supplementary Figure S11.** Structure library of the AuNP-functionalized small origami rings to calculate the averaged TEM image (see Figure 2b). Scale bar, 50 nm.

AuNP-functionalized small ring -  
after rotation

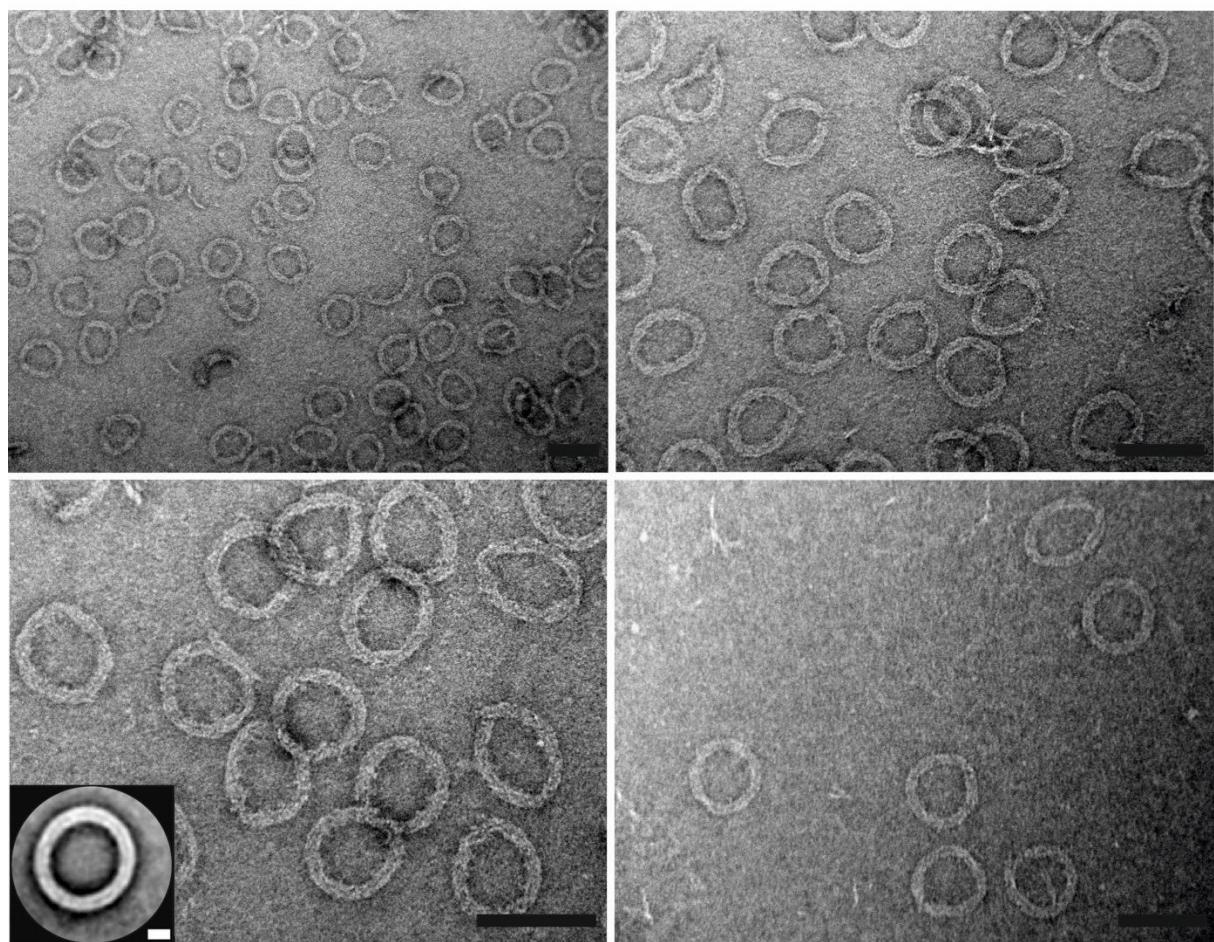


**Supplementary Figure S12.** TEM images of the AuNP-functionalized small origami rings after the rotation process and classification of the structures. The structures are divided into intact (black) and defective (red) ones. Some structures might have been defective directly after the structural assembly, but not from the rotation process. Scale bar, 100 nm.



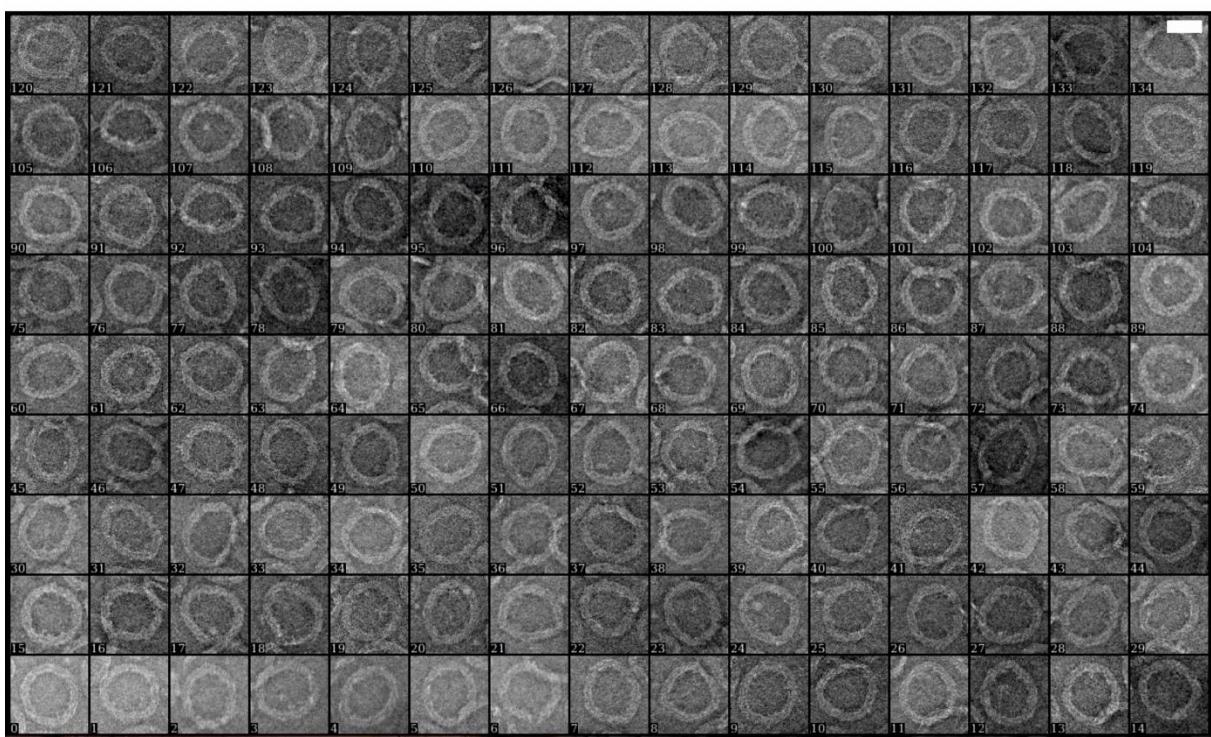
**Supplementary Figure S13.** (a) Bidirectional rolling of the AuNPs along the small origami ring (for exact angle changes see Supplementary Figure S4). (b) Experimental fluorescence data. (c) Calculated fluorescence data.

Large ring



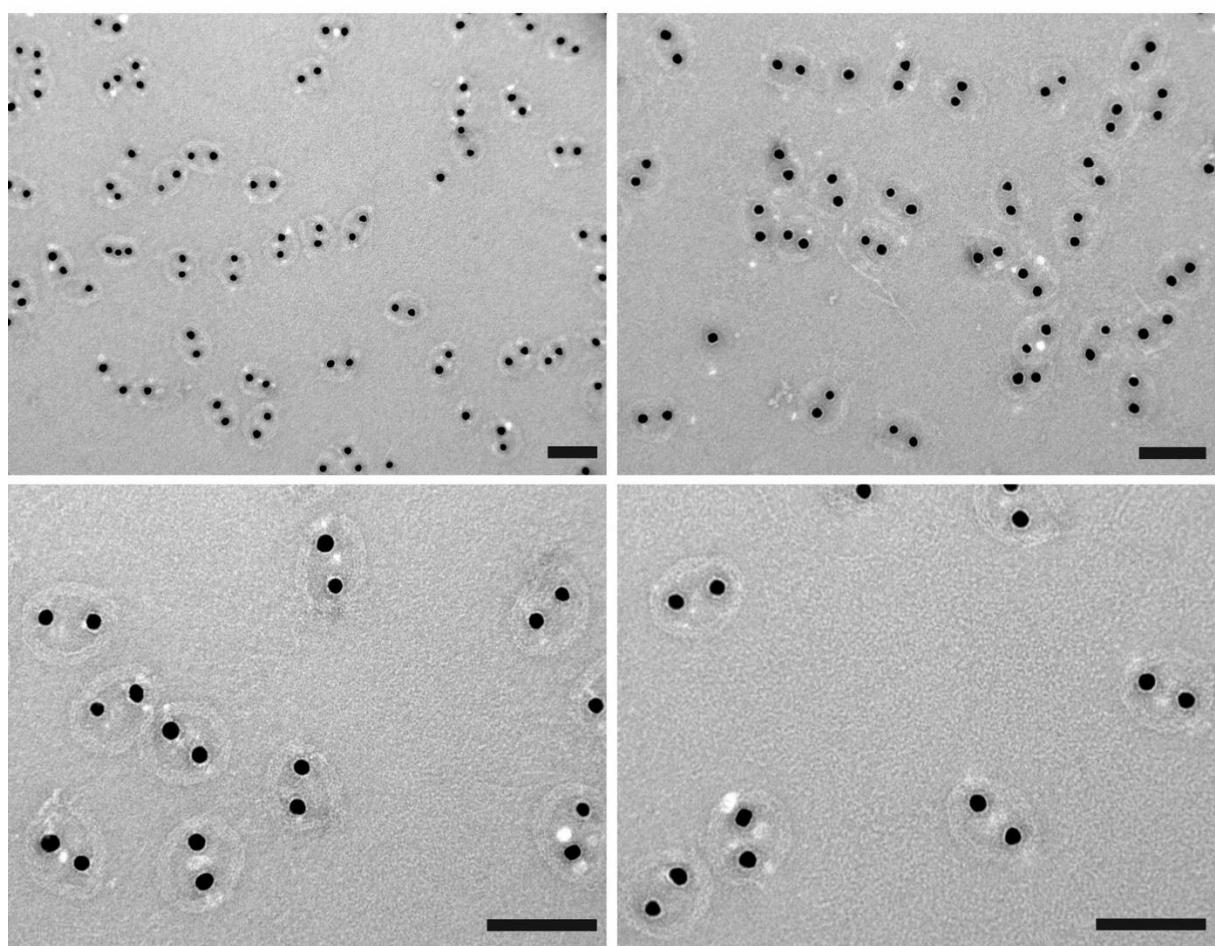
**Supplementary Figure S14.** TEM images of the large origami rings. Scale bar, 100 nm. Inset: averaged TEM image (see Supplementary Figure S14 for the structure library used for the calculation). Scale bar, 20 nm.

Large ring



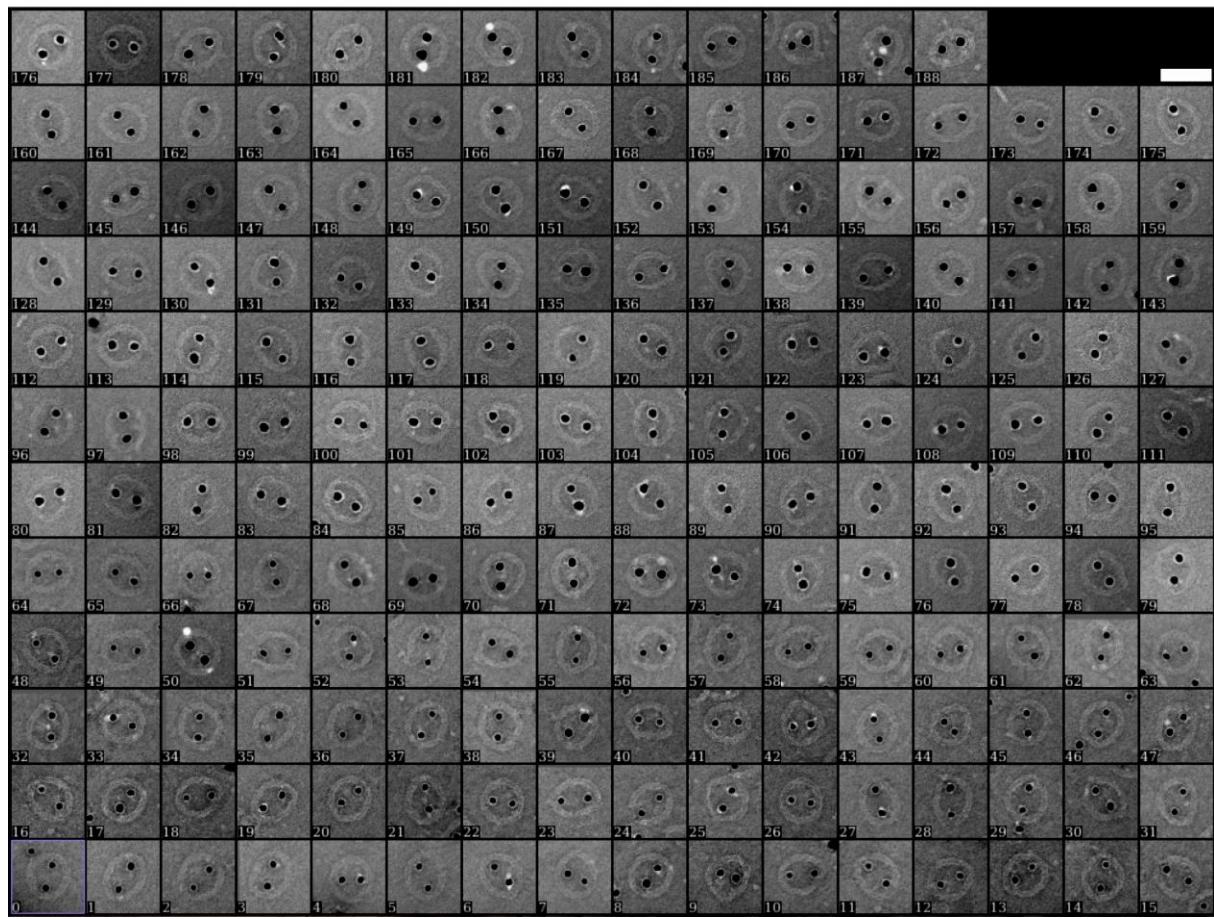
**Supplementary Figure S15.** Structure library of the large origami rings to calculate the averaged TEM image (see Supplementary Figure S13). Scale bar, 50 nm.

AuNP-functionalized large ring



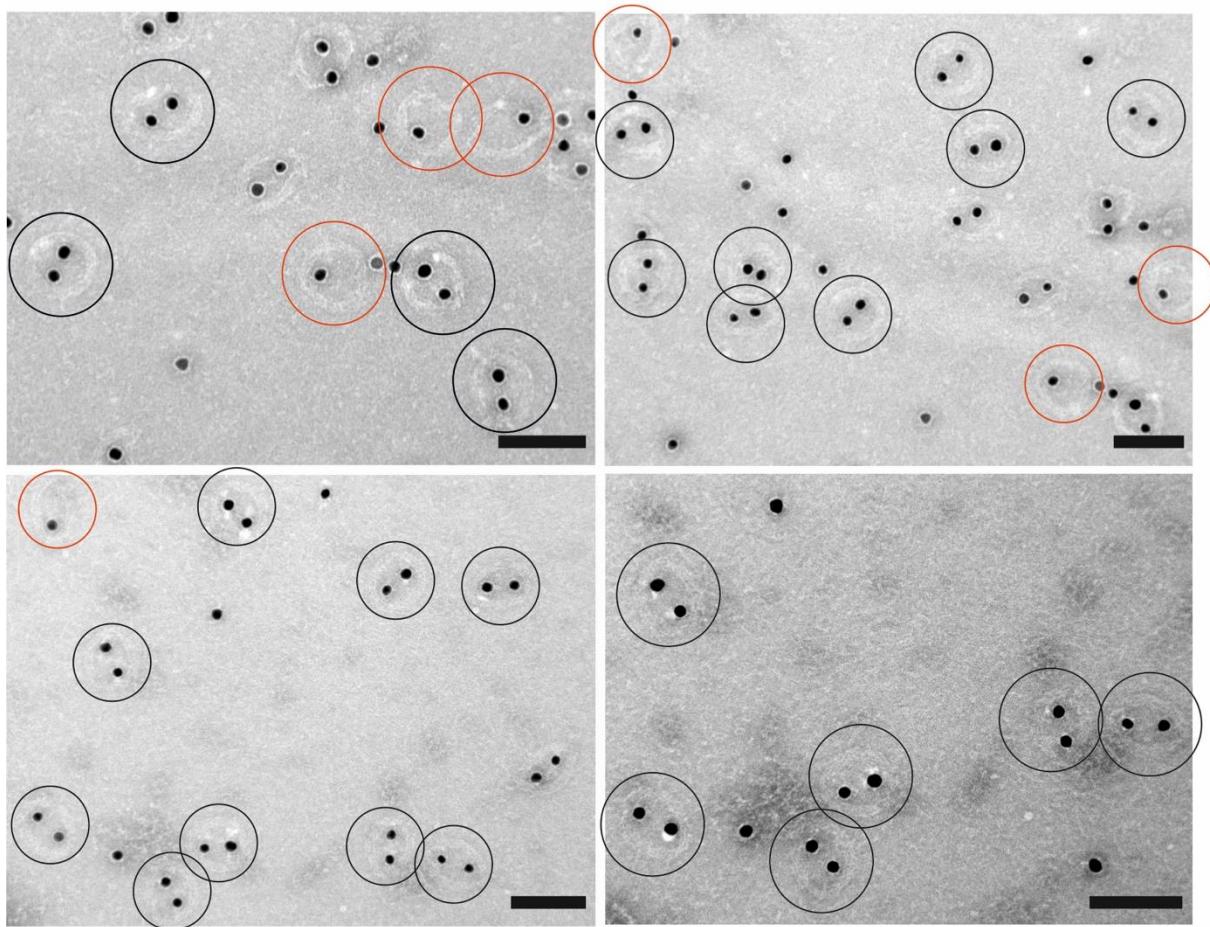
**Supplementary Figure S16.** TEM images of the AuNP-functionalized large origami rings. Scale bar, 100 nm.

### AuNP-functionalized large ring

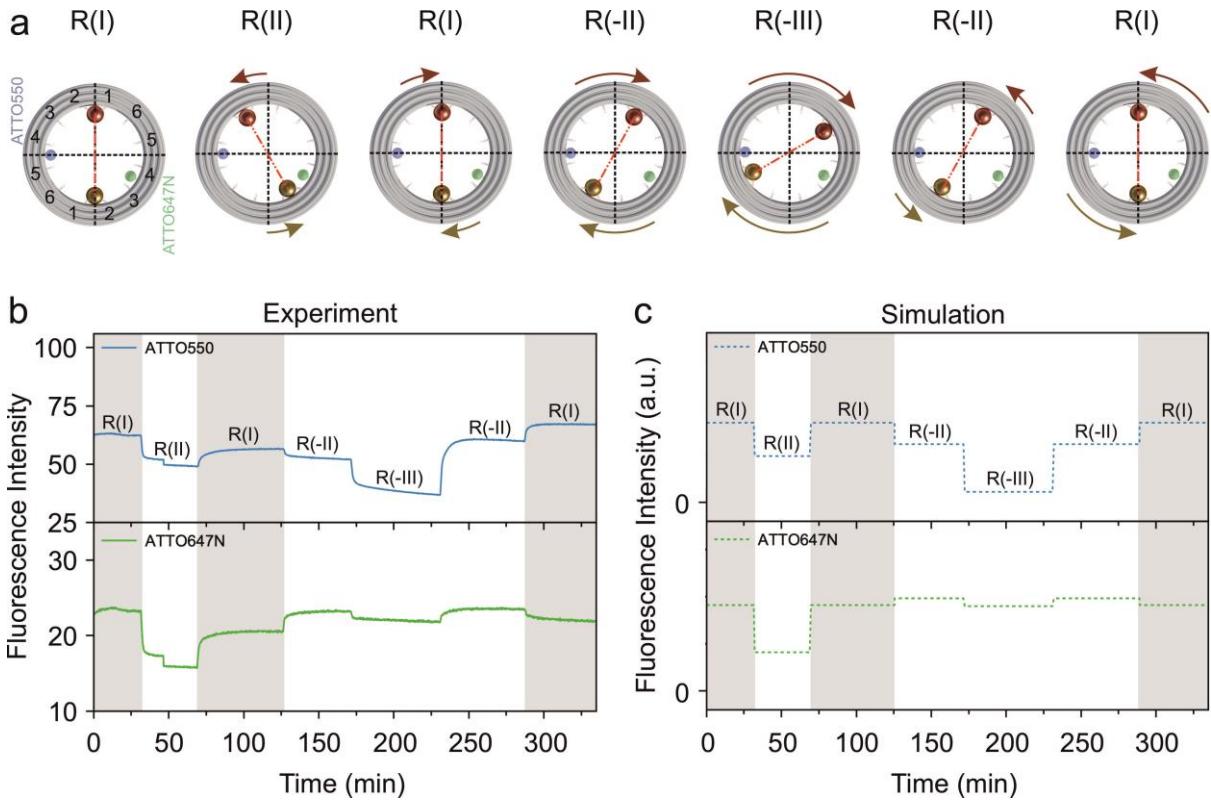


**Supplementary Figure S17.** Structure library of the AuNP-functionalized large origami rings to calculate the averaged TEM image (see Figure 3b). Scale bar, 100 nm.

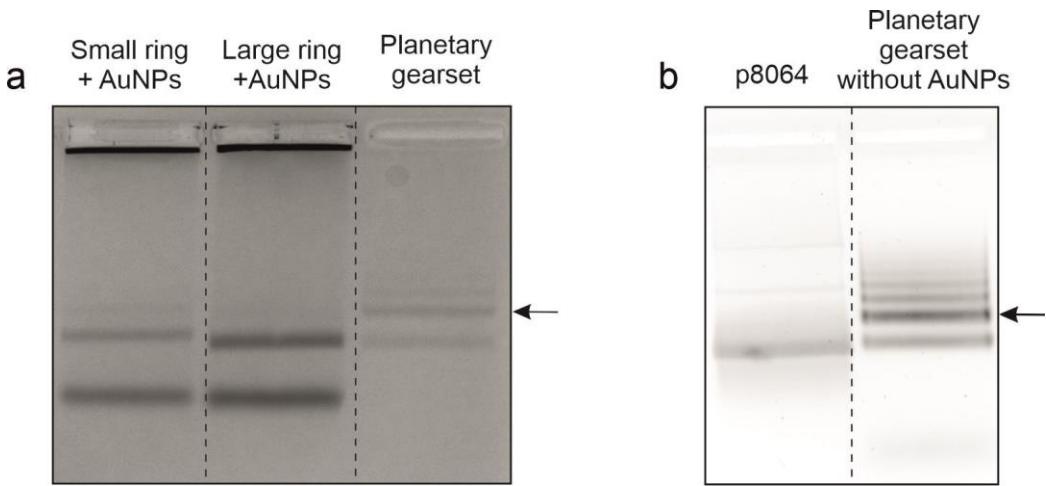
AuNP-functionalized large ring -  
after rotation



**Supplementary Figure S18.** TEM images of the AuNP-functionalized large origami rings after the rotation process and classification of the structures. The structures are divided into intact (black) and defective (red) ones. Some structures might have been defective directly after the structural assembly, but not from the rotation process. Scale bar, 100 nm.

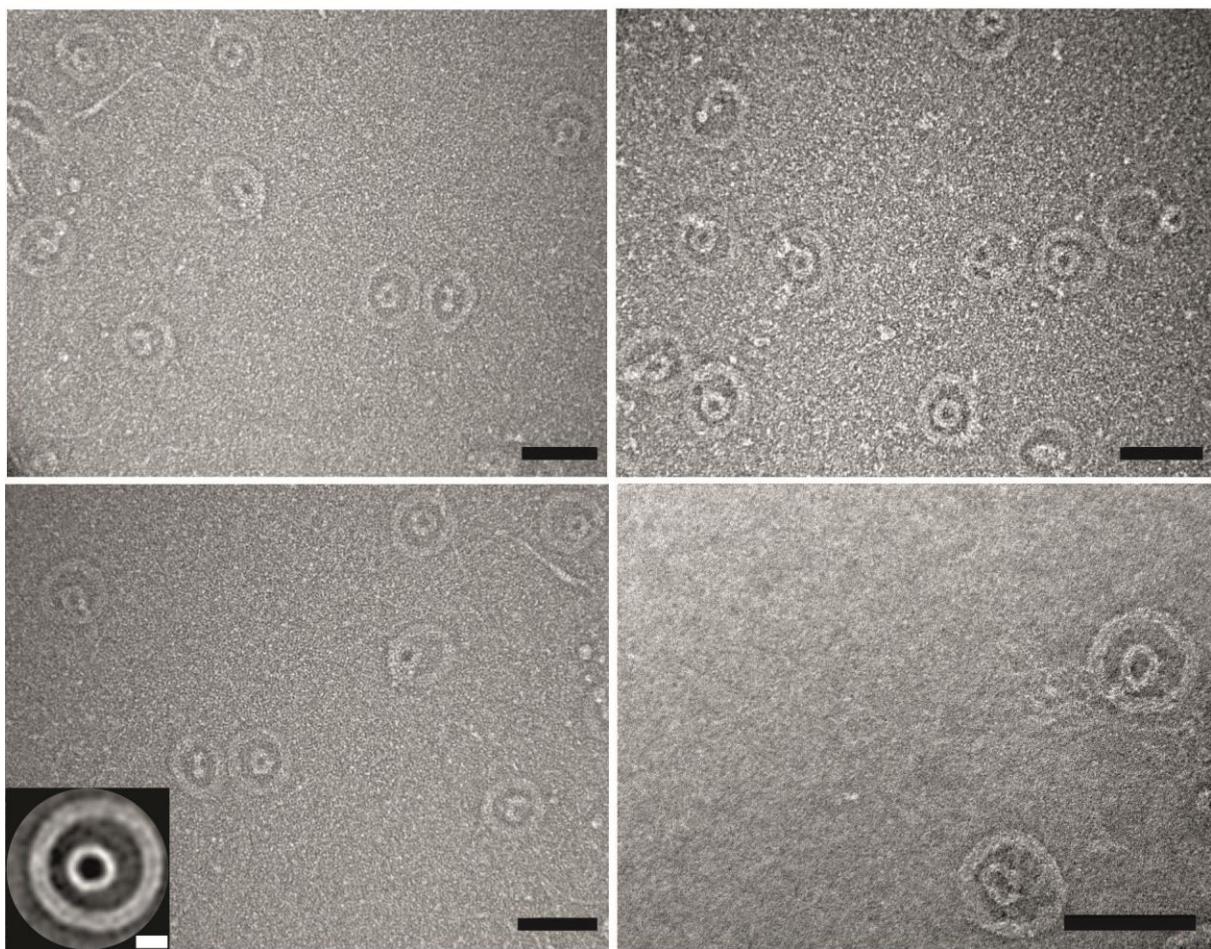


**Supplementary Figure S19.** (a) Bidirectional rolling of the AuNPs along the large origami ring (for exact angle changes see Supplementary Figure S3). (b) Experimental fluorescence data. (c) Calculated fluorescence data.



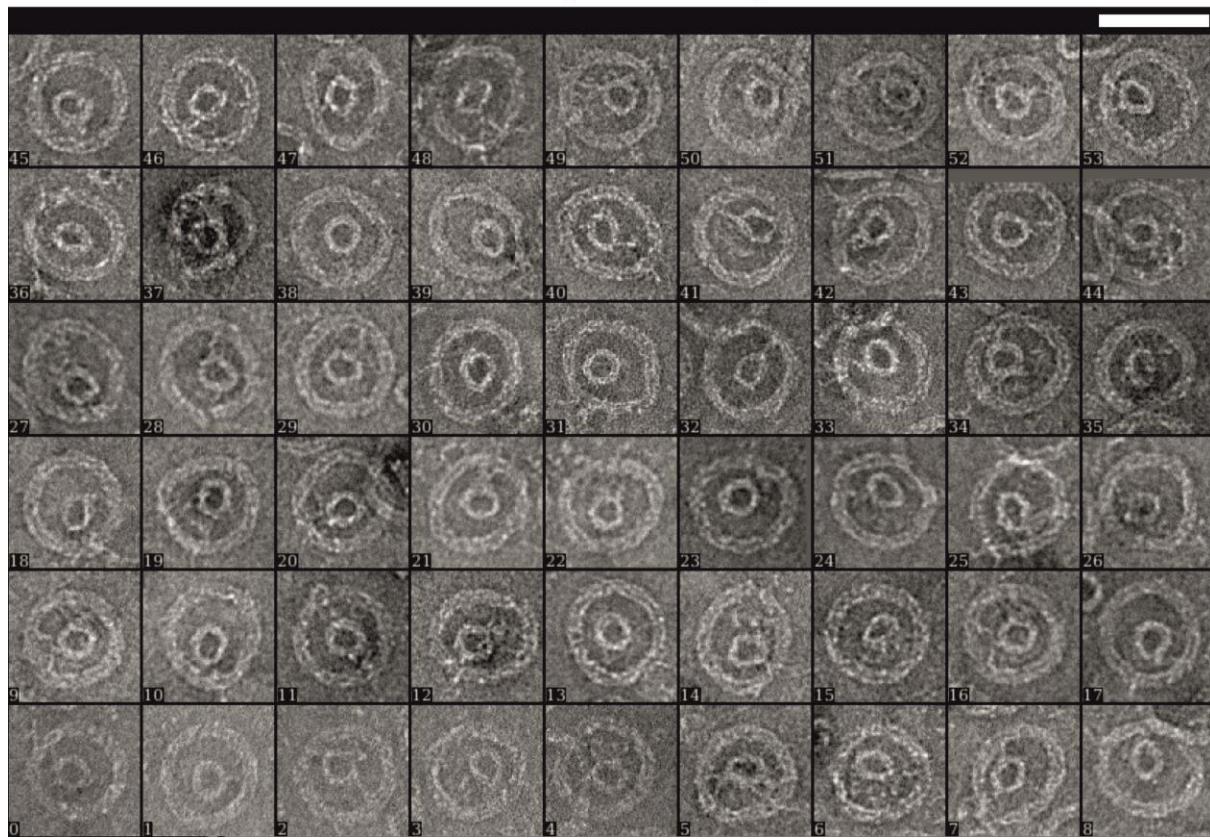
**Supplementary Figure S20.** (a) Agarose gel electrophoresis image of the assembled rotary nanodevices. The AuNP-functionalized small and large origami rings are used as reference. The band marked with the arrow contains the desired nanostructures. (b) Agarose gel electrophoresis images of the small and large origami rings connected only by the locking strands. The band marked with the arrow contains the desired double ring structures. The p8064 scaffold is used as reference.

Locked large and small rings



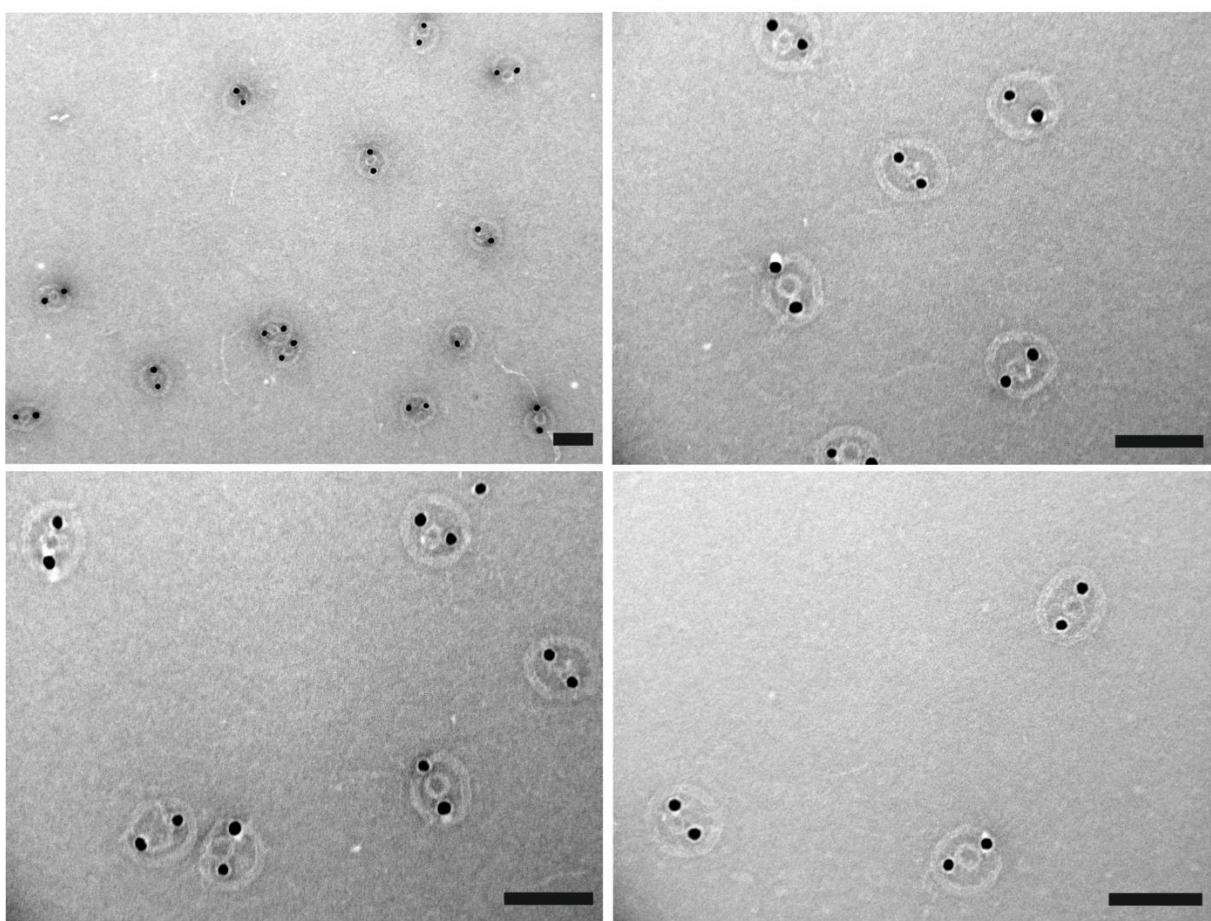
**Supplementary Figure S21.** TEM images of the double rings connected by the locking strands only. Scale bar, 100 nm. Inset: averaged TEM image (see Supplementary Figure S20 for the structure library used for the calculation). Scale bar, 20 nm.

Locked large and small rings



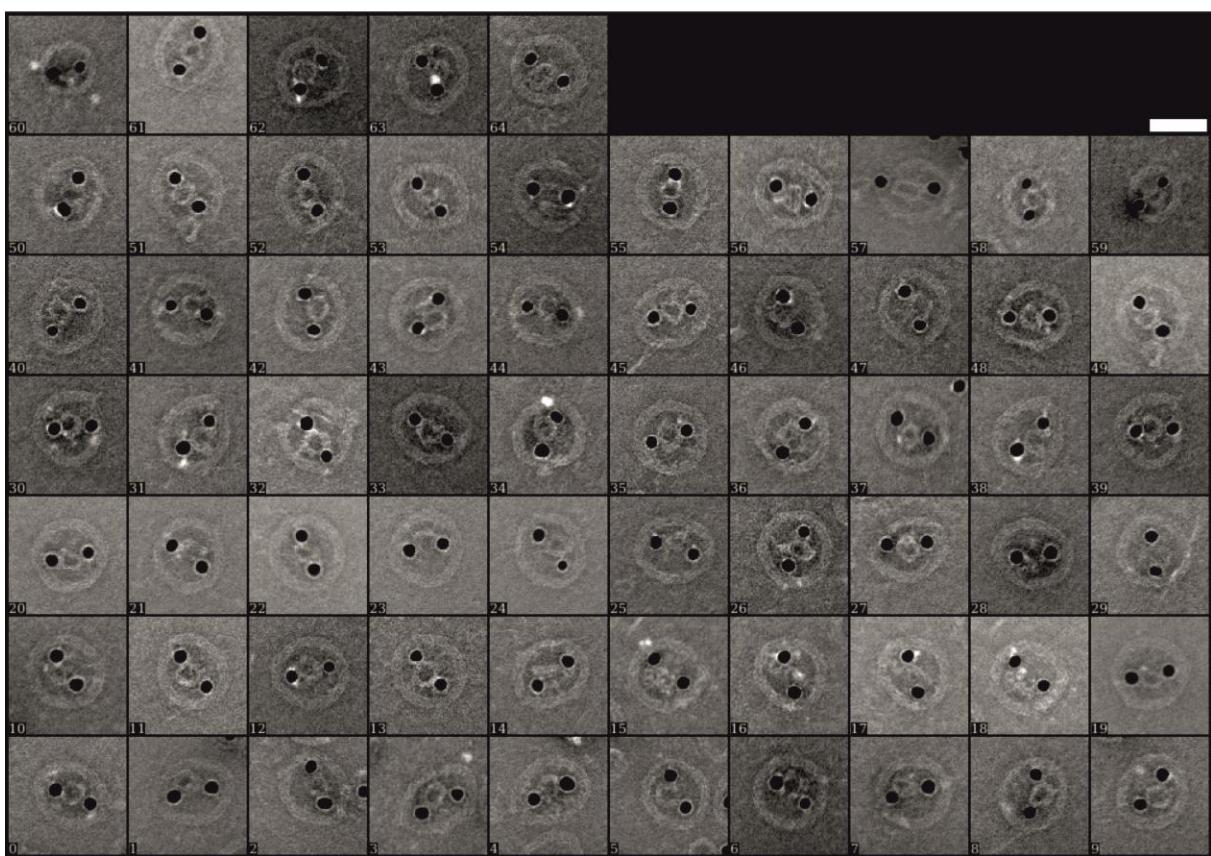
**Supplementary Figure S22.** Structure library of the connected double rings to calculate the averaged TEM image (see Supplementary Figure S19). Scale bar, 100 nm.

Planetary gearset nanodevice



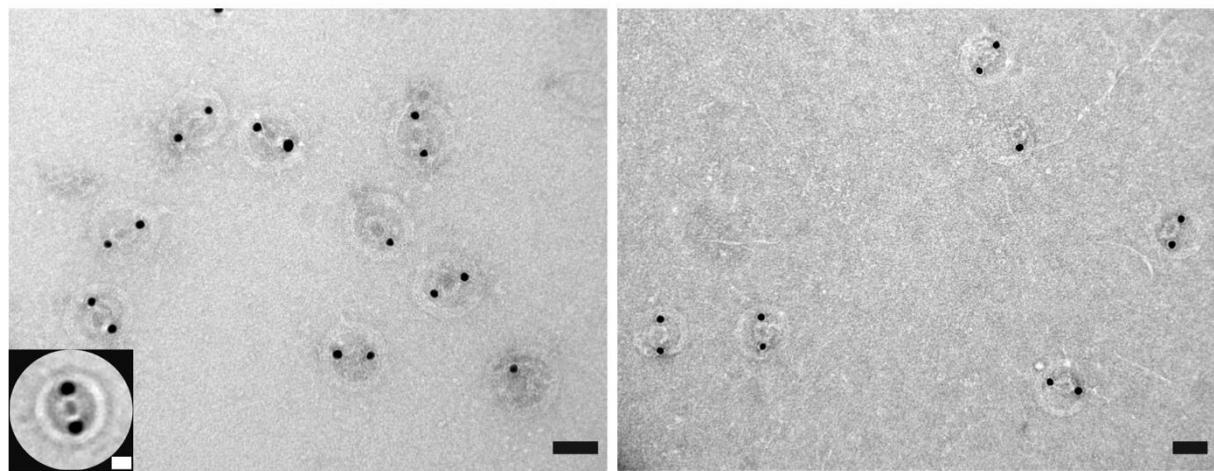
**Supplementary Figure S23.** TEM images of the assembled planetary gearset nanodevices. Scale bar, 100 nm.

Planetary gearset nanodevice



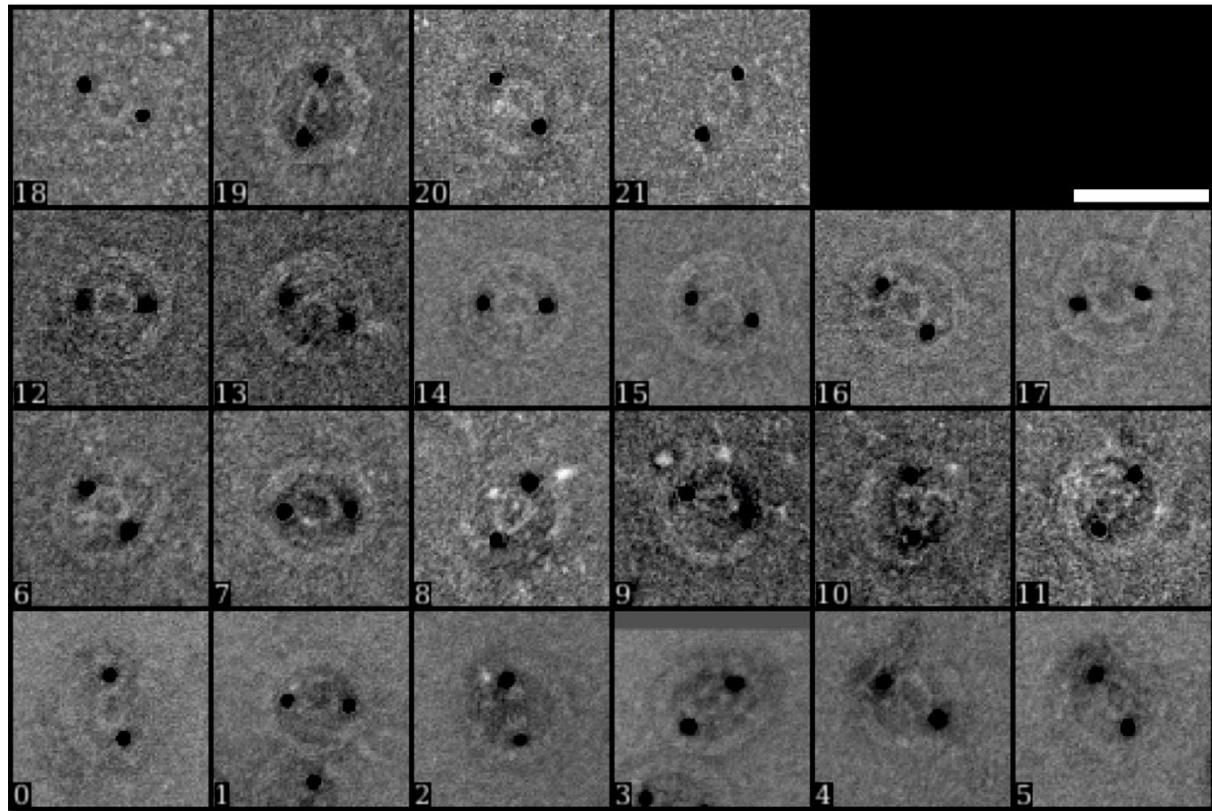
**Supplementary Figure S24.** Structure library of the planetary gearset nanodevices to calculate the averaged TEM image (see Figure 4b). Scale bar, 100 nm.

Planetary gearset nanodevice - 10 nm AuNPs

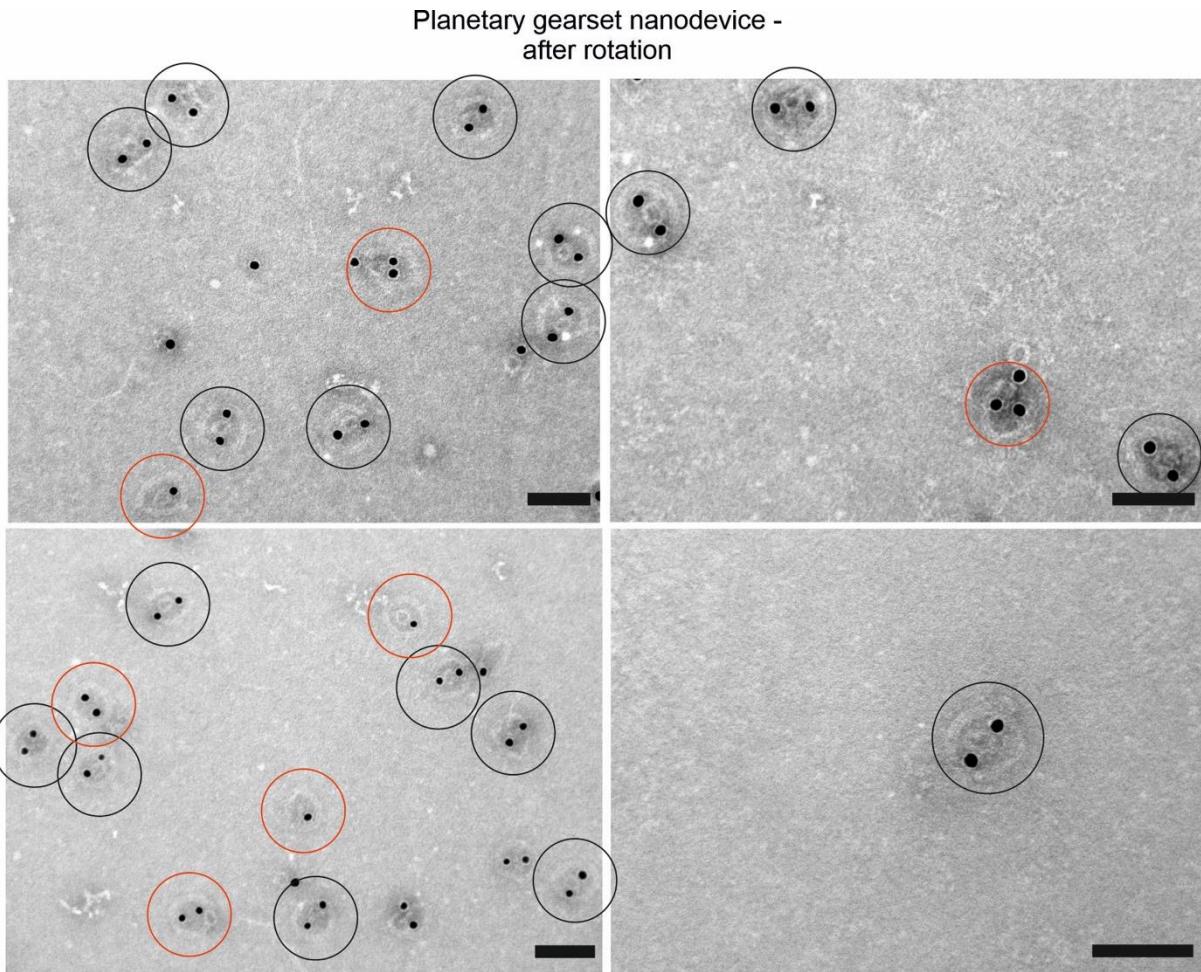


**Supplementary Figure S25.** TEM images of the planetary gearsets with 10 nm AuNPs. Scale bar, 100 nm. Inset: averaged TEM image (see Supplementary Figure S26 for the structure library used to calculate the averaged TEM image). Scale bar, 20 nm. Due to the less tightly-fitted geometry, the small rings show weaker deformations on the TEM grid after the deposition and drying processes.

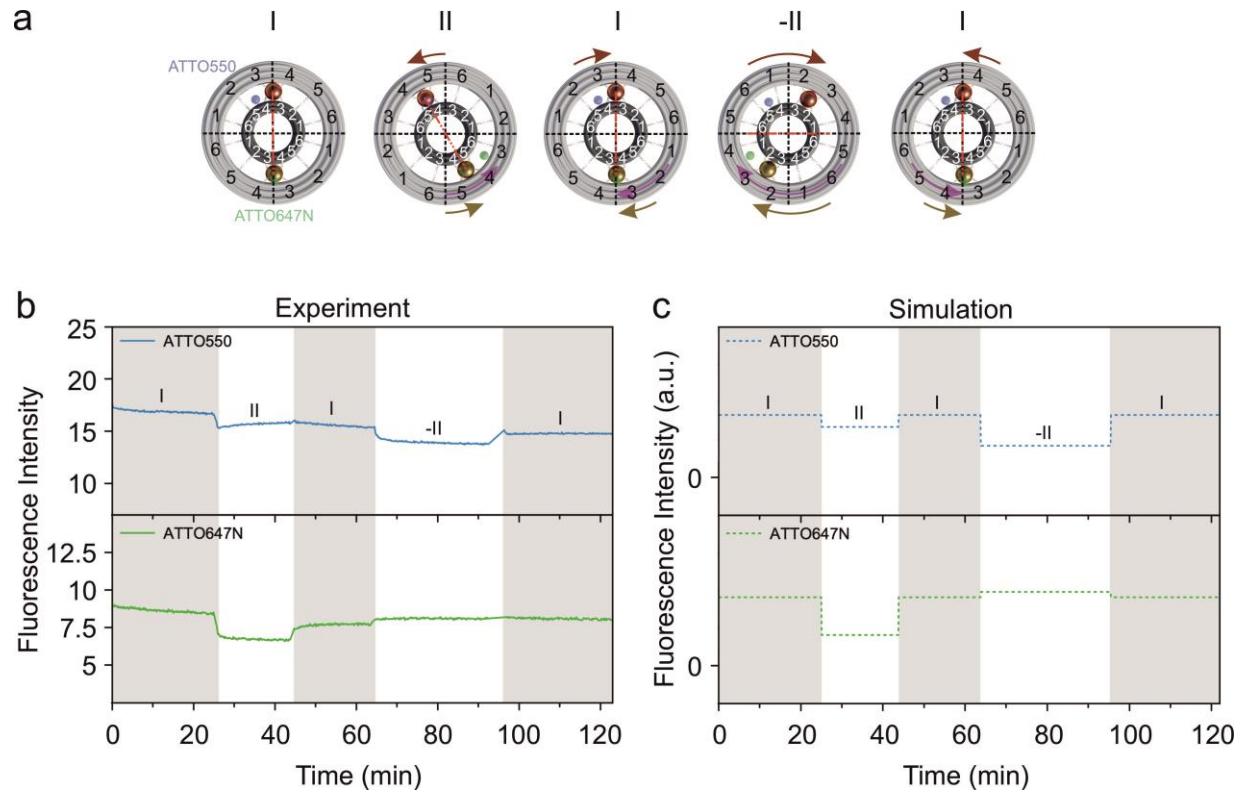
Planetary gearset nanodevice with 10 nm AuNPs



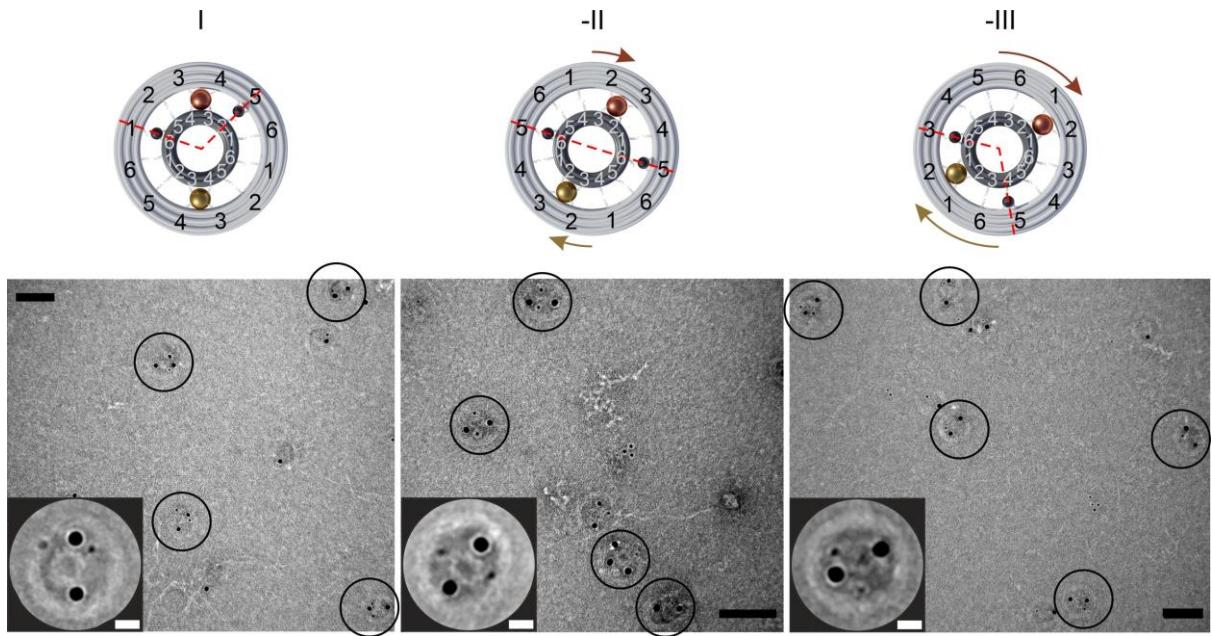
**Supplementary Figure S26.** Structure library of the planetary gearsets with 10 nm AuNPs to calculate the averaged TEM image. Scale bar, 100 nm.



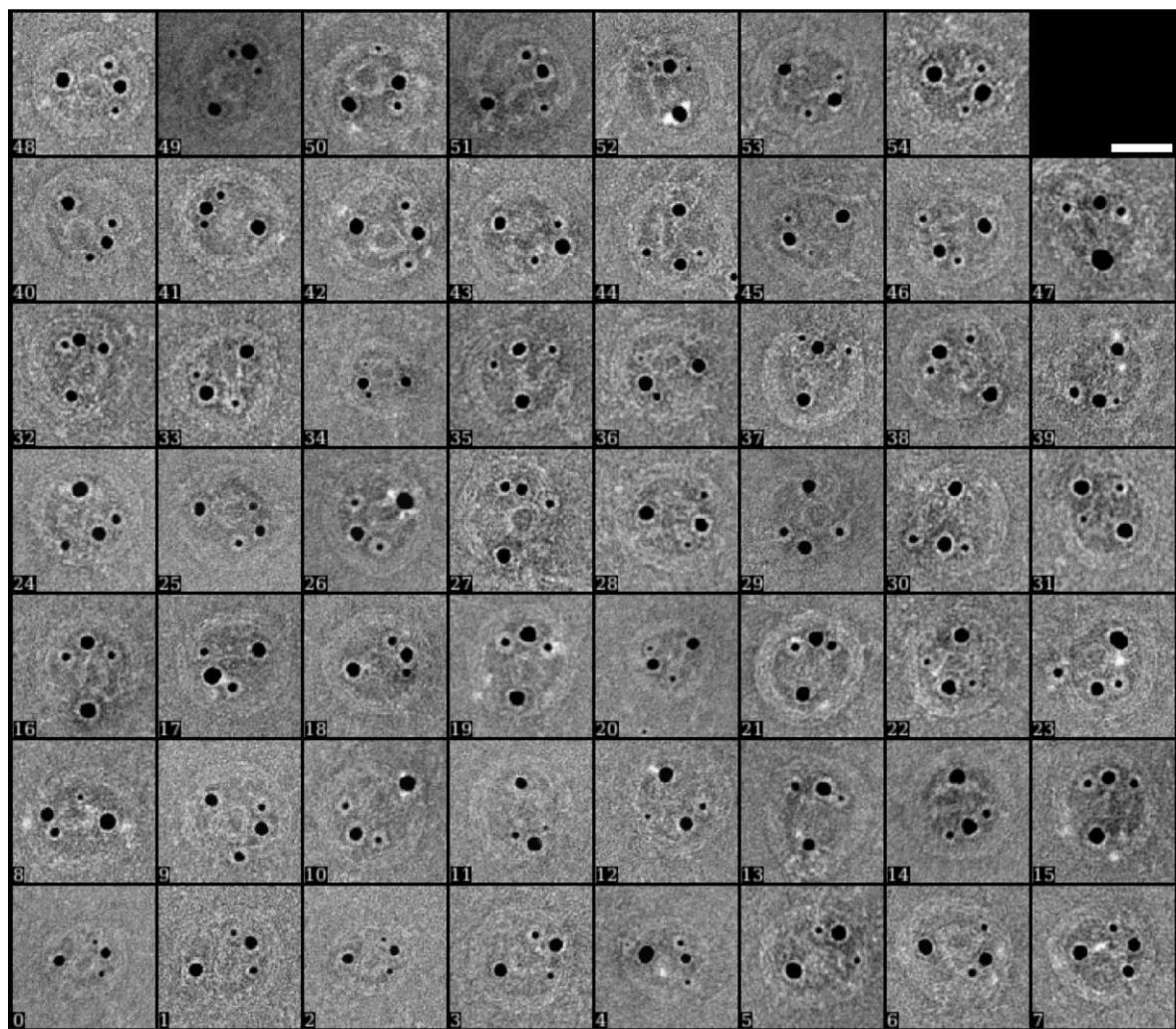
**Supplementary Figure S27.** TEM images of the rotary nanodevices after the rotation process and classification of the structures. The structures are divided into intact (black) and defective (red) ones. Note that some of the defective structures might have been formed directly after the structural assembly but not be due to the rotation process. Scale bar, 100 nm.



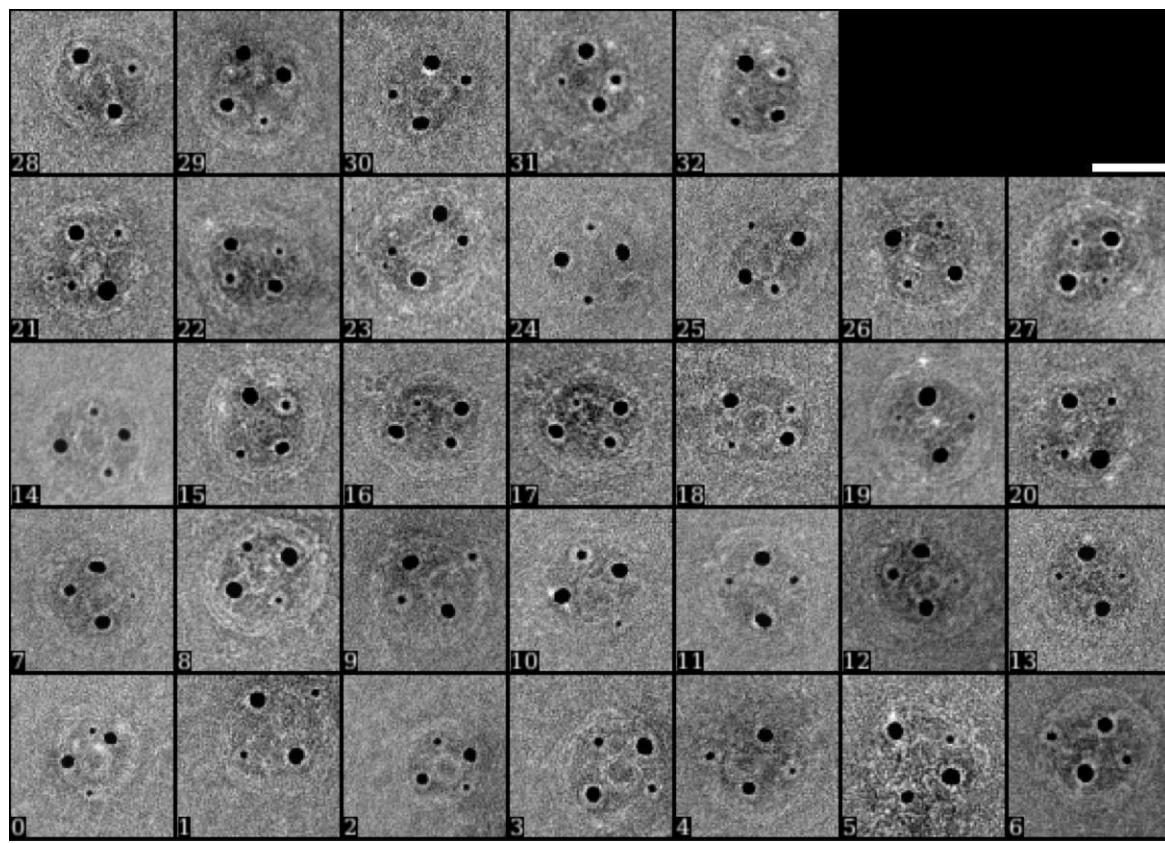
**Supplementary Figure S28.** (a) Bidirectional sliding of the AuNPs in between the small and large origami rings (for exact angle changes see Supplementary Figures S3 and S4). (b) Experimental fluorescence data. (c) Calculated fluorescence data.



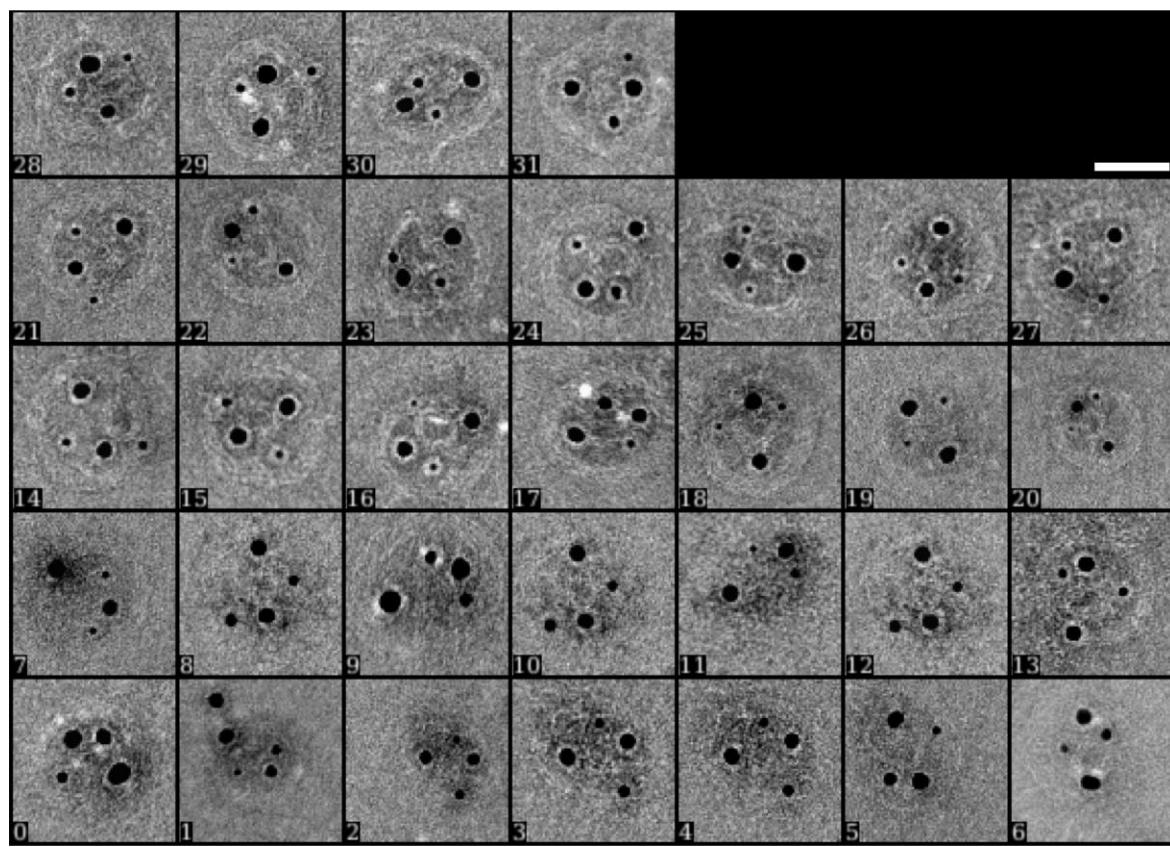
**Supplementary Figure S29.** Additional AuNPs as markers to validate the rotation mechanism. One AuNP is immobilized at foothold row 6 on the small ring, while the other AuNP is positioned at foothold row 5 on the large ring. Schematic of three representative states ‘I’, ‘-II’, and ‘-III’, in which the two new AuNPs (5 nm) are illustrated using black balls. The gearset is driven from state ‘I’ to ‘-III’ and the corresponding TEM images at these states clearly prove the designated relative movements among the AuNPs, which agree well with the predictions based on our rotation mechanism. Scale bar, 100 nm. Inset: averaged TEM images (see Supplementary Figures S30 to S32 for the structure libraries used to calculate the averaged TEM images). Scale bar, 20 nm.



**Supplementary Figure S30.** Structure library of the planetary gearset nanodevices at state 'I' to calculate the averaged TEM image (see Supplementary Figure 29). Scale bar, 50 nm.



**Supplementary Figure S31.** Structure library of the planetary gearset nanodevices at state ‘-II’ to calculate the averaged TEM image (see Supplementary Figure 29). Scale bar, 50 nm.



**Supplementary Figure S32.** Structure library of the planetary gearset nanodevices at state '-III' to calculate the averaged TEM image (see Supplementary Figure 29). Scale bar, 50 nm.

## Supplementary Tables

**Supplementary Table S1.** Samples added to drive the rotation for real time fluorescence detection for the small rings in Figure 2. The blocking and releasing strands were prepared at a concentration of 400 µM in H<sub>2</sub>O. Generally, small volumes but high concentrations of the blocking and releasing DNA strands were used to power the AuNPs to reduce the dilution effect. The total volume increase was 4.5 µL (4.3%) after the rotation process.

Step	Strands added	Volume added per strand
‘r(I)’ to ‘r(II)’	1, $\bar{3}$	0.25 µL, 0.25 µL
‘r(II)’ to ‘r(III)’	2, $\bar{4}$	0.25 µL, 0.25 µL
‘r(III)’ to ‘r(IV)’	3, $\bar{5}$	0.5 µL, 0.25 µL
‘r(IV)’ to ‘r(V)’	4, $\bar{6}$	0.5 µL, 0.25 µL
‘r(V)’ to ‘r(VI)’	5, $\bar{1}$	0.5 µL, 0.5 µL
‘r(VI)’ to ‘r(VII)’	6, $\bar{2}$	0.5 µL, 0.5 µL

**Supplementary Table S2.** Samples added to drive the rotation for real time fluorescence detection for the large rings in Fig. 3. The blocking and releasing strands were prepared at a concentration of 400  $\mu$ M in H<sub>2</sub>O. To reduce the dilution effect, small volumes and high concentrations of the blocking and releasing DNA strands were used. After the rotation process, the total volume increase was 4.5  $\mu$ L (4.3%).

Step	Strands added	Volume added per strand
‘R(I)’ to ‘R(II)’	1, $\bar{3}$	0.25 $\mu$ L, 0.25 $\mu$ L
‘R(II)’ to ‘R(III)’	2, $\bar{4}$	0.25 $\mu$ L, 0.25 $\mu$ L
‘R(III)’ to ‘R(IV)’	3, $\bar{5}$	0.5 $\mu$ L, 0.25 $\mu$ L
‘R(IV)’ to ‘R(V)’	4, $\bar{6}$	0.5 $\mu$ L, 0.25 $\mu$ L
‘R(V)’ to ‘R(VI)’	5, $\bar{1}$	0.5 $\mu$ L, 0.5 $\mu$ L
‘R(VI)’ to ‘R(VII)’	6, $\bar{2}$	0.5 $\mu$ L, 0.5 $\mu$ L

**Supplementary Table S3.** Samples added to drive the rotation for real time fluorescence detection for the planetary gearset devices in Fig. 4. The blocking and releasing strands were prepared at a concentration of 400  $\mu$ M in H<sub>2</sub>O. To reduce the dilution effect, small volumes and high concentrations of the blocking and releasing DNA strands were used. After the rotation process, the total volume increase was 4.2  $\mu$ L (4%).

Step	Strands added	Volume added per strand
unlocking	Open locks L1, L2, L3	0.2 $\mu$ L
‘I’ to ‘II’	3, $\bar{5}$	0.2 $\mu$ L, 0.4 $\mu$ L
‘II’ to ‘III’	4, $\bar{6}$	0.2 $\mu$ L, 0.4 $\mu$ L
‘III’ to ‘IV’	5, $\bar{1}$	0.4 $\mu$ L, 0.2 $\mu$ L
‘IV’ to ‘V’	6, 2	0.4 $\mu$ L, 0.2 $\mu$ L
‘V’ to ‘VI’	1, $\bar{3}$	0.4 $\mu$ L, 0.4 $\mu$ L
‘VI’ to ‘VII’	2, $\bar{4}$	0.4 $\mu$ L, 0.4 $\mu$ L

**Supplementary Table S4.** Distances between the fluorophores and the respective adjacent AuNPs used in the theoretical calculations for the large origami ring.

State	ATTO647N	ATTO550
	Distance to AuNP center (nm)	
‘R(I)’	25	30.5
‘R(II)’	16.5	23
‘R(III)’	12.5	15.5
‘R(IV)’	16	12.5
‘R(V)’	24.5	16.5
‘R(VI)’	32	25
‘R(VII)’	25	30.5

**Supplementary Table S5.** Distances between the fluorophores and the respective adjacent AuNPs used in the theoretical calculations for the small origami ring.

State	ATTO647N	ATTO550
	Distance to AuNP center (nm)	
‘r(I)’	34	30
‘r(II)’	26	26
‘r(III)’	16.5	16
‘r(IV)’	12.5	12.5
‘r(V)’	16.5	16.5
‘r(VI)’	26	22
‘r(VII)’	34	30

**Supplementary Table S6.** Distances between the fluorophores and the respective adjacent AuNPs used in the theoretical calculations for the rotary nanodevice.

State	Large ring – ATTO647N	Small ring – ATTO550
	Distance to AuNP center (nm)	
‘I’	12.5	16
‘II’	16	12.5
‘III’	24.5	16.5
‘IV’	32	22
‘V’	25	30
‘VI’	16.5	26
‘VII’	12.5	16

### III. DNA Sequences

**Supplementary Table S7.** DNA sequences of the core staples for the small origami ring.

Core-1	TCCGGGAGAAGCCAATCAGAAAAGTAT
Core-2	GTCACGACGTTGTCCCCGGTCATAGCTGTT
Core-3	CTCATGGGTACCGAGCTCGAAAAATTGCATT
Core-4	GGTCAATAGGGTTGATAACCTAGCATTATGCCTGCAGGT
Core-5	TTATCCGCCCTATAAAATAGCGGTCGCCCTGGCCCTGAGATTGC
Core-6	ACGTCACTTTGCGGATCAACATGTTAAACA
Core-7	TAATTCGAGCTCAAAGCG
Core-8	ACCGCGTTATTATAGTCAGAACGAGAACGAGAATGATAT
Core-9	AAAAACCGTCTACATTCAACTAAGAGGAAGCAGGGCG
Core-10	TTGGAACAAGAGTCCACTATTAAAGA
Core-11	GCTTAATTGCCCTTACCCCTGACT
Core-12	TCCGAAATCGGGCCCCAGGGCACACAGCTGATTGCTTCTTCACCAGTG
Core-13	AGAGGTTGCAGCACAAAGTTGTGTGAAATTG
Core-14	CTACGTTAATAAAACGAAGAGAACGATGATGCTGAGAGTAGTAAAA
Core-15	GTATACTGCCAGGGCGATCGGCCAGTGCCAAGCTGCAACCTGCGCAA
Core-16	AAACTAGCATGTCACGTTGGGAAGTT
Core-17	TTGTATCAATAGGAACGCCGCCAGCTACGTTGGT
Core-18	AATATTAAATTATCAGCTATTGTGAGCGCGGATTGA
Core-19	TAACCGGAAGCATAACAACGTGATGGTGGT
Core-20	TTGTTATCGTAAAATTCCATGAGCTAACTCACATTCTATTACGCCA
Core-21	TTTAGAACCTCATATACTGAAAAG
Core-22	AAATTATCGATTAAGTTGGTAACGCCAGGGTTCCAGCTGG
Core-23	TCATCAATTCTACTAAAGGCAAGGGCAAAGC
Core-24	GAGACAGTCAAATAACAGTTGATT
Core-25	GTACAGGGTAGCTATTGCGCATCGTGTCTGGCCT
Core-26	ATTAAGCAATAAGCCTCAGAGCATAAAGCAGATCGCA
Core-27	TTAGCAAAGTGGCATTGGGCGCGAGTTAAATGCAATGC
Core-28	TATTACAGGTAGATCATA

Core-29	TCACCTCAGGATAAATCGGTTGTACCAAAGGATAAAATGT
Core-30	CTGGAAGTTCATCCATATCACCATCAATATGACCATA
Core-31	TCCTGTAATCAAAAAACAGGAAGA
Core-32	TCCTCGCAAATGGTCAACCATTAGATACATAATAGCCCAGGGATTAG
Core-33	GTAAGTAAAGATTATTCAACGCAAAACATTGACGACGA
Core-34	CGACTCTAGAGGATAAAACGACGGTGC GG
Core-35	GCTATACGAGTAGATTAGTTGAGTGTTGTGCAA

**Supplementary Table S8.** DNA sequences of the fluorophore-modified DNA strands for the small origami ring. The fluorophores are attached to a dsDNA sequence (blue).

ATTO550-a	[ATTO550]ATGGAACGCAGGTCTCATCACATTAAATTAAACAAG CA
ATTO550-b	GAGACGAGGCATAGTAAGATGGATAGCCTATCAGGATGCGTTCC AT
ATTO647N-a	[ATTO647N]ATGGAACGCTCAGGATCCAATAATCATACTAGTAGT GTTTA
ATTO647N-b	CCAACAAACCTGTCGCATGCGTTCCAT

**Supplementary Table S9.** DNA sequences of the foothold staples for the small origami ring. The foothold region with AuNP-capturing sequence is highlighted in green. Footholds 1c and 4d have two functions, which additionally provide the double-stranded stem region for respective locking strands (red).

Foothold-1a	TCTGAATGGATTGATCA CTCCAGCCAGCTTCCGGCACCGCT
Foothold-1b	TCTGAATGGATTGATCA AATCAAAAATCAGGTTGAATATAAT
Foothold-2a	TCTGAATGGGTACTGTGT CAGTATCGGACC GTCTAGCTGATCTTAAAC
Foothold-2b	TCTGAATGGGTACTGTGTAGTCAGAAAAGCGGAGACTCAAATATG TGGACGGCCGCTGAGTAAAAT
Foothold-3a	TCTGAATGGTTAACAAACCATCTGCCAGTGCCGGAGCCGGTTGATTC AAAAGGGTGAGAAATCAA

Foothold-3b	<b>TCTGAATGGTTAACAAACCTGAATCCCCTAATGCAGATAACATAACGCC</b> AAAACGTCATAAAATATTCAT
Foothold-4a	<b>TCTGAATGGAGTTGTTGAGTAGATGGGAGAGATCTACAAAGGGTCC</b> AATA
Foothold-4b	<b>TCTGAATGGAGTTGTTGACTGCGGAATGGAATTATTTAGGAATACCA</b> TCAGGAATCACCCCCAAATAAT
Foothold-5a	<b>TCTGAATGGAAATCCTGTCCGTAATGGGTCAATTGCAACGGTAATCGT</b> A
Foothold-5b	<b>TCTGAATGGAAATCCTGTTAGACGCAACACTAAAGATTCA</b> GAAAAAT
Foothold-6a	<b>TCTGAATGGTCAAGTGTAAACGGAGTAACAACCCGTCGGTTGTTAAG</b> TAAACGT
Foothold-6b	<b>TCTGAATGGTCAAGTGTACCCTCGTTGGGGTAATCTGGAGCAAC</b> A
Foothold-1c	<b>TCTGAATGGATTGATCAGTGCTGCAAGGTCTCCGTGGAAAAAGTG</b> TAAAGCCTAAAGA <b>AGTGA</b> CTACGA <b>ACTAGACCTAGTATCGATGACT</b>
Foothold-1d	<b>TCTGAATGGATTGATCATAAAACCAAAATAGACGCAGGCGAAA</b> TCCTGTTGAACAAACAT
Foothold-2c	<b>TCTGAATGGGTACTGTGTCGAAAGGGGCTAATGAGCACAACATACG</b> AGTATT
Foothold-2d	<b>TCTGAATGGGTACTGTGTAGCGAGAGGCTTGCAGGGTGC</b> GAT
Foothold-3c	<b>TCTGAATGGTTAACAAACCGCCTTCGAATTGCGTTGC</b> GTCTGGC GCC
Foothold-3d	<b>TCTGAATGGTTAACAAACCAAGGGTGGTC</b> CTTCACCCACGCTGGTT AAAATCTCA
Foothold-4c	<b>TCTGAATGGAGTTGTTGACTGTTGGAGC</b> TTCCAGTCGGGAGCGCG GGG
Foothold-4d	<b>TCTGAATGGAGTTGTTGAAGAGGCGGTGATAC</b> TTAATTGCTC TTGCGG <b>ACTACGA</b> ACTAGACCTAGTATCGATGACT
Foothold-5c	<b>TCTGAATGGAAATCCTGTGCCATTG</b> CCTGCCAGCTGTATGCAACTAA AGGGAAACCAAAAGAA
Foothold-5d	<b>TCTGAATGGAAATCCTGTTAATGAATATAAGAGTCCA</b> A ACA
Foothold-6c	<b>TCTGAATGGTCAAGTGTTC</b> CTGGCCTACGGTGTCCAATTCTGC GA ATT
Foothold-6d	<b>TCTGAATGGTCAAGTGTG</b> CTGTAGCTGGCTTAGAAACCAGACCGGA ATCCAGT

**Supplementary Table S10.** DNA sequences of the locking strands for the small origami ring. The double-stranded stem region is colored red and the locking sequences are highlighted in orange.

Linker-1'a	<b>AGGTGAGTAAGT</b> TTT <b>AGTCATCGATACTAGGTCTAGTCGAG</b> GGATGACCCTGTAATACTTTGGCAACCGTGC
Linker-1'b	<b>AGGTGAGTAAGT</b> TTTAGTCATCGATACTAGGTCTAGTCGAGTCAA TGAAATTAAATTGACTACGAACTAGACCTAGTATCGATGACT
Linker-2'a	CGTCAACCGAAATTGCATCAAAAAGATCTC <b>GACTACGAACTAGACCT</b> <b>AGTATCGATGACT</b>
Linker-2'b	<b>GTACTTAGTGT</b> CTTAGTCATCGATACTAGGTCTAGTCGAGTCTTG CCAGAACCAAGACGACGA
Linker-3'a	<b>TGTAGAGGATGA</b> TTT <b>AGTCATCGATACTAGGTCTAGTCGAGTCTCA</b> GGATCCAATAAATCATACTAGTAGTGTGTTA
Linker-3'b	<b>TGTAGAGGATGA</b> TTTAGTCATCGATACTAGGTCTAGTCGAGTCCC ACAACCTGTCGATGACTACGAACTAGACCTAGTATCGATGACT

**Supplementary Table S11.** DNA sequences of the core staples for the large origami ring.

Core-1	CGGCCAGAACATCAGCCTGTGCACTCTGTAGCATCAGCGGGTCAGACGAT
Core-2	CAGCACAGACGATTGGCCTAACAGGGAGGT
Core-3	TTGCTCGTCATAAACATCTGCATTA
Core-4	TCACCCCTGCTGGTCTGGACCAGCAAGTTCT
Core-5	CTCAATCAGGAATTATGAAGATAAAACACAGTGCCA
Core-6	CGGATTGCGGGAGAACGCCTAACACCTGTAATACATCTAACAT
Core-7	TATACCCCTAGAACATCCTGAGACGCTATTAA
Core-8	GCAACACCAAAACATTATGATTAAAGCTA
Core-9	ATGCCAAATAATTGCGTCTGACAATAGGA
Core-10	CGCTGAACAAAGAACCCACATAACATTAT
Core-11	GTATAGGTAATCGTTGATAAGCATGCAATGCCTGGTGAGA
Core-12	CATTTGCAAATTAAATCCTTGC
Core-13	CAGCATATCATATTCCGAAGAGAAGGAGCGGAGAAGAAG
Core-14	GCTGATAAACACATGTTCTAAACCAGACGACGACTCAGGGC
Core-15	AATTGAGGACTAAAGACTTCGAACGGCTA

Core-16	AAGGCCAGCTTCATCAACACACGGCCTCCTGTAACAGCAA
Core-17	CCGAACGTTATTAAATTACTTGAATAAAAACAATTGACAACTCGTCC
Core-18	CGGAACTGAGTTCGTCACCGAAACCCATG
Core-19	AGAGCGGTTACCAGGCCAAATCAATAGA
Core-20	CAATATTTGTTAACGTACACAATAAGAAACGAGGTTTG
Core-21	CTTTAACTCAATCCGCCGGGATCATTGCAG
Core-22	ATCGAATTATCCAATCCAAAAAAATAAA
Core-23	CACCAAATAAACCTCATTGTTAGATATTACAAACTCCCTC
Core-24	GCATCAGGTGTGTTAGTTGAGCTACGTGAACCA
Core-25	GCGCGTTAACGAACTGGCGTACATTACCTATGCTGAATCA
Core-26	ATAGTAAGATTAAGAGGAAGCTAAAGCGGA
Core-27	CGGGGCGCCTGTAGCATTCTGAGAGTACAAACTACCACGCCA
Core-28	GTAATACATTCAACCGATTGGAAAGACAAAAGGGGGAAACG
Core-29	CGTAACCGGAATTGTGAGTTATCATGTTACCAGGGCAGGG
Core-30	AGTTAGTAAAGTAATTCTGTCGATAAGTA
Core-31	CTTCCTCCATTAAACGGGTGATTTCATGAGGAATTAGGCT
Core-32	CACTCCGAAACGTACAGCGCCCTCACCGG
Core-33	GCGACCTTAATCATTGTAATGAGATGGT
Core-34	GGCTGGAGGTGTCCGGGTACACCGCGTCGGTGGGCCATCCCACG
Core-35	ACATGTTAATTGCTTAGATTCAATAACCTTCATTGAATCC
Core-36	AGGAGCCGGTGAATGTAACAGTTCAGCCGAATAAT
Core-37	AAGGAATTCAACTACTTTGCAATGTTAGGGATATTCAATT
Core-38	AGACTCCTCAGTACAATCAGTGCCTTGAGCCTATT
Core-39	GTAGATGGTAATGGCGCCAGCGGA
Core-40	CTAGCTGAATCACCAGCAAACGCATGTCAAGTGGCATCAA
Core-41	GGAATTGGGAGTGACAGCTGCCGATAGTTCAGGAGGTTT
Core-42	GCGTCAGTAGCCCCCAGTAGCACCATTAAATCGATAG
Core-43	GCCCCCTGTAACAGTAGCGGGCGAGAGGGTCCCTCAGAGC
Core-44	CATTACCCCTATCCTATCGGCTGTCTCCGCACTC
Core-45	TGAAACCCCATTAGGGCATTCTTTCATATACGCACTATG
Core-46	ACCGACCACCGGAAAATGCAAATCCAATTATATT

Core-47	ATTGTGTACGGAGACCAACTGGCGCATAGATATATTGGT
Core-48	GCCCTTACCAAGAACGTGAGCGCTAATAACAATAATA
Core-49	CCAAGTATCCTTATTCAAGATAGGCCTTACCAACGCTCA
Core-50	GAGGCATTTGAGCTTAGCAAGCACCATTAAACAACGCCAACAT
Core-51	TTTCAAACGCAAGAAAATAAGGCCTGTTATAATTACATT
Core-52	CGCCATATTAGCGTACCAAGAGGGATCACCGTCACGCCAAAAT
Core-53	TTAACGTATAAAGAAAGTTACTTACCTGAGATAACATTGAG
Core-54	AGTACGGCGATAACCGGAATCTCATACATGGCTTTAA
Core-55	TGAGTGAATAACCTCTAATAAGGCCTTTAATGGAAACAGTACAAC
Core-56	TTAGCAAACCGAGGCTGGCATGGCGCATTAGACTCAGAGG
Core-57	TTCTAACATTGCCATTGGCGATTAGAGAGTATTTTG
Core-58	ACCCAAACGGTGTATCAAGAGGTAAAGAATACACATACCAA
Core-59	GCGCTTCGCCAACCGAGCTTAC
Core-60	AAACAATCGAAGTGCCAAGCTTC
Core-61	ACGCCATCAGAAATTGCGATTAAA
Core-62	AATCGGTTACAAGGCAAAGAATT
Core-63	TTGCATCAGAAAATCAAAATCAG
Core-64	TTAATTTCAGCTTGCCCTGACGAG
Core-65	CAGAGGCTCAGTCACCCCTCAGCAG
Core-66	TACCGTAAGCTCAGAGCCACCACC
Core-67	TGAGGCAGTTAGCCGCCACCAGAA
Core-68	AAATTCAATCGAAAGAAACGCAAAG
Core-69	CAGCCATAGAAACGCTAACGAGCG
Core-70	CCGACAAAAATGTAATTAGGCA
Core-71	ATTAATTATCAAATCAATATATG
Core-72	AGCCGCACATCGACGTCTCGTTAACGGAACCTAACGGT
Core-73	GCCTCAGCAGTTGGCTGCGCTCGCTACAGGAAGATT
Core-74	TTGCTGGTAATATCCAGAA
Core-75	AGAGCCACGACTTGCAAAATCAGG
Core-76	TGCTATATTCTAAGTGCAGGATAAGAAAAATAATAACCAAT
Core-77	AGAGGTGGAGCCGCCGTCCGTTTGACGTTGAAACGACGGCCAA

Core-78	TATCAGGTCGATGAGCCGGAGACAGTCATAAAATTAA
Core-79	TTTTGTTAAATCACATTGCCATTAAACGTTAATATTTGTTAGC
Core-80	TCTACAAAAGGAGAGGGTAGCTATTAAAGCCTCAGAGCATCAACCGTT
Core-81	AGCAAAATTAAGCAATTGAGAGTCTCATCCAATAATCATACAGGAC
Core-82	ACGATAAGTTGCCAGGAATACCACATTACGAGGC
Core-83	GTCTTACCTGACTGGCTCGAACGAGAAAACGAGAATGACCATCG
Core-84	GGTCAATGGACAGAGAAAACAAAGTACACGAAATCC
Core-85	AAACACCAGAACGAGATATAGCGAGAGCTCATTCACTGAATAAGGAC
Core-86	TAATAACATCACTGAAAACAAATGTTAGGTAAAATCATAGGT
Core-87	CGAAAGACAGCATCTAACCGAACTAGGCCGCTTGCAGGATCAA
Core-88	AAGGCTCTTTCACGTTGAAAATAGCAAGCCCAATAGGACAACACTAAA
Core-89	CTCATTTCAGGGATCTTATCGGTTCACCTCAGAACCGCCACCCCTG
Core-90	GATTGGAAGGTTAATAGATTGGAAC TGATAGCGAACCCAC
Core-91	CCACCACCAGAGCCCAAGGATTAGGT CAGAGCCACCACCTCAGTA
Core-92	TATTAGACTTACCCAATTGCGTACCTAC
Core-93	ACACCACGGAATAAGACCGCTTTCACATAAAGGTGGCAACATATAAA
Core-94	ATCTTACATAAGAAACAATGAAATAATTGCCAGTTACGAATTGAG
Core-95	TCTTCCAGAGCCTAGAGTAAGCAGTTATCCTGAATCTTACCAT
Core-96	ATCGTAGTTAACAGCAAGCAAGCCGTAGTAATAAGAGAATGGTATTAAA
Core-97	CGCTGCTAACACAGACAACCAATTCCAGACGTTATAAACAA
Core-98	CACCCAGCTACAAATGATAACCCACCCCTG
Core-99	TAGAAAATACATATCCAAGGCCGGGAATT
Core-100	TTAATTGAGAACATCGAACATTCCAAGCGAGC
Core-101	CCCTCAGAACCGCATGAATAGAACATGGG
Core-102	TGCAGGGAGTTAAGATTGTATCACCCCC
Core-103	AGAACCGCCACCCATTGCCCGTAGTACTG
Core-104	GTCCATCCGACTAACCGCGATAGAACGGCGCCAATTATTTTC
Core-105	ACAGTATAATTACTTACAAATAACCTTTAACTGTAAAT
Core-106	CTTTAACAGTTAGGAATATAATTGATA
Core-107	AGTAGTAGCATTAAGGATCAATATGAAGAT
Core-108	AGTTGATCCATTAGTTGGCTAGAGCTTTAAATAT

Core-109	ACGTAACAAAGCTGGATGCAGATAGAAAG
Core-110	ATATTAAATTGTCAAGGGGACGGTTGGT
Core-111	TTTCCCAGTCACTCATAAAAAATCTGCT
Core-112	TTTGAATTACCTTTCAAAGAACTTATAT
Core-113	GGGGAAAGCCGGCCCCCGGAAAACTAAAGAGAACATTGCCTTGAGAGA
Core-114	TTGCTTGACGAGATGACACTGATACTTCGATTTAATTGCCAAAAAA
Core-115	CCAACGGCAGCACGGTCCCCCTTAACG
Core-116	TAAAGTTTGTGCGTCTGCCACTAC
Core-117	CATTGCGGATCAAGCCGGACCGAGGTGCCGT
Core-118	CCAGCGCGTGCCGGGGTTCTAGCCTCCTCAGCAGCAAC
Core-119	TGGGAAAGTCATTATACCATAATCTCACCCG
Core-120	CATATCAATAATGGAAAGAACGAACTATCGGCC
Core-121	AACTATACTCCGGCATCATATTAG
Core-122	GTAATAATTGATGATATAAGGGA
Core-123	ATTTGCGTAAATGCCGACACACGTATAACG
Core-124	AACAAAGGGGAGAAATACCCATAATCAGTGAGG
Core-125	ATGTAGAATCCCATGAACCTCCACGCAAATT
Core-126	TCAAAAGGAGTAATATATGTAGAACGTGGCGA
Core-127	CTGAGAGACTCGCGCCTGT
Core-128	CCAGGCATGGGAAGTCCC GTGCATCTCGAAGATCG
Core-129	ATTCAACACAATTGGATAGGCT
Core-130	TTCCGGCAGCCTCCATGGCGGTTGTACAGGCGGC
Core-131	TTAACGCCTCAGAGAACGCCAAATAAACGTCAAGATTAGT
Core-132	GGATT CGCGCAGAGAGCGTAAAACAGAACAGATGAA
Core-133	AACGCCGATCGGTGAAGGGGGCTGCGGATTGACC GGCGCAT
Core-134	TCCACAGGCGGGCCCGCGTGCCTCTGCAGCCAGCGAGTGTAC
Core-135	CCGGGCAGCCAGCTTCCGGCTATTAAACCAGTATCG
Core-136	GTATTAAGAGCACCTATTATTCTGAAAGCCGCCAGCATTGACAGTTAAT
Core-137	AGCGATTAAAACATGGCTGAGCC
Core-138	GCCGTCCGTAATCAGTAGCGTTATTGTACAACGTCACCAA
Core-139	ATTGCGTGATGAAGGGTAAAACGGGAACGGATAACCGTAAAAAA

Core-140	TTTTATTTGCCCGAAAGACTTCAAAATG
Core-141	GGGGTGCCTGAGAGAGTTGCAGCAAGCGGTCCACGCTGTCTGGCCAAC A
Core-142	CCAAAAAGAGCAACACTATCTAGTAAATTGGGCTAACGCCAA
Core-143	AGACGGGGCAAACATAGCGATGCTTC
Core-144	GGAGAAACAATCAGTAACAGTACCTTTAAAAGTTGAGTTTCAGG T
Core-145	TGCGCCATTAATTGCCTGCCTAATGAGTG
Core-146	CAGCAGGAAGCCTAGTGTACGAAATCCCTTATCCGCTCACAAAT
Core-147	AGCTAACTGCCAACATACGAGCCGGAAGCATAA
Core-148	CACTGCGGCCGCGTTGCGGTGGGAATG
Core-149	TAATGGTTGAGATTCATCTTCTGACGCTTGTAAATCGTAAAAACTT
Core-150	GGAAAGTCCGCTCACTGCCGCTTTCCTGTGTGAAA
Core-151	TCATTCCATAGTTAAAGTACGGTGTCTATTATAGTCAGAAGCGTAGCTC A
Core-152	TCAGGAGAGCCAGCAGCAAAT
Core-153	TCCTGAACAGCAGCCTT
Core-154	GTCAATAGATACAAAGGGTATATAATCCTGA
Core-155	TTGTATCCGCCTGGTTTCTTCACCAGTGAGACGGGCCCTGAAAGCGT
Core-156	AACAAGAATTATTCAAACAAACATATACTGGA
Core-157	AAGGGAAAAGATAGACTTATGTGCCTAAATCGGAACC
Core-158	CCCTCGAGGGGTAATACTGCAAGAACTAAAC
Core-159	CGCCTGATACAGGAATTACCGTTCCAGTAAGCG
Core-160	CGCAACCAGAGCACACGTCACTGCAGAGAGA
Core-161	GCTAACACAGGAGGCCCGGAAC
Core-162	AAAGCATGCAAGGCGAGTACTTATAGCTCTACGG
Core-163	GCGCGTAACCACCATGACAAGAACCACTATTACGAAAAATCTAC
Core-164	TCTTCGCCGGGTACAGCCCTACACTGATGCCGGTTACTCCAGTCG
Core-165	ACACGAGCATCATCTAAACTAACACCGCCTGCAA
Core-166	GAGAATCAGTAATAAAAGGGACATGTTGCC
Core-167	GCCCGAGATCGCTCAATCGTCTGAAATCAGGCGGTAAAAAAT
Core-168	TCACCTACATTGAAAGGGTTGAG

Core-169	AACGTCCCACATTAAAGAACAAAAACGC
Core-170	GAAAAACCTGCTGTCAGTTGGGATTATTACATTGGCAGTGGTTCCGA
Core-171	TGTTGTTAAGAATAATCGGCAAAATCCTGTTGATGGATTCAACCAGTC
Core-172	TACGTGGCACAGGTTGCGTATTGGCGCCAAGAGTAAAGGGCGAAA
Core-173	ATTGCCCTCAAAATCAACCAGTTGGATCATGGTCATAGCTGGT
Core-174	AAGTTGTTAGAACCCCTCTGAAACAGCTG
Core-175	GGGAGAGGCGACAATATTTTGACAAA
Core-176	GAGGCGCAACTGCTCCATGTTACTGGAACGAGGGTAGCCCTGATAA
Core-177	CCGCGCTTAATGCCCTCACAGACCATGAAAGACATAAGGGGCCGGAAC

**Supplementary Table S12.** DNA sequences of the fluorophore-modified DNA strands for the large origami ring. The fluorophores are attached to a dsDNA sequence (blue). The complementary sequence to hybridize the ATTO550-a strand is provided by Foothold-4d.v2 (see Supplementary Table S13).

ATTO647N-a	[ATTO647N]ATGGAACGCAGAGTAACAACCCGTTCATTAATGTGATCAGAACTTGAC
ATTO647N-b	ACCCTCATATATTTA GCGTTCCAT
ATTO550-a	[ATTO550]ATGGAACGC GACGGATTGAGGGAGGGAAAGATTAAATCCTGAGAAGTG

**Supplementary Table S13.** DNA sequences of the foothold strands for the large origami ring. The foothold region with the AuNP-capturing sequence is highlighted in green. Footholds 3, 4 and 5d, e and f have a two-nucleotide thymine extension (dark orange) placed between the foothold sequence and the DNA origami structure. Foothold 4d has several functions. It serves as a linker (red: stem section of the locking strand; orange: locking sequence; purple: toehold sequence) and provides the hybridization sequence for the ATTO550 dye (Foothold-4d.v2, blue), respectively.

According to whether only the large origami ring or the entire rotary device is assembled, the addition of foothold strands has to be adjusted.

Foothold-6a	<b>TCTGAATGGTCAAGTGTATGAATTGCCTGTCGTGCCGAGCTCTA</b>
Foothold-6b	<b>TCTGAATGGTCAAGTGTAAACCGTCGAATTGTAAACAGGGCGGCCAACCGCGC</b>
Foothold-6c	<b>TCTGAATGGTCAAGTGTTCAGGGCGATGCCAGATCCGTCCGTGCCAGCATTTCACGTGCTG</b>
Foothold-5a	<b>TCTGAATGGAAATCCTGTCGTAATGGTAAAGATTGCCCTGCGGGCGTGGAAATCAAGTTTT</b>
Foothold-5b	<b>TCTGAATGGAAATCCTGTAAAAAACAGCCGCCAGCAGTGGAGCCGGT</b>
Foothold-5c	<b>TCTGAATGGAAATCCTGTTGGGTTGTAGATCCTCACGCTGGCAACACGGTAAACGATGCTG</b>
Foothold-4a	<b>TCTGAATGGAGTTGTTGAATTCTCCGTGGAACAAACGCCGTGGTG</b>
Foothold-4b	<b>TCTGAATGGAGTTGTTGACTAAAGTGGCGACGGCCTCAACTGTAAGCGCCACCGCTCTGGTG</b>
Foothold-4c	<b>TCTGAATGGAGTTGTTGAGCCCCGATTAGAGAACGCCCCAAATAGATAGGCGG</b>
Foothold-3a	<b>TCTGAATGGTTAACAAACCAAATTGGTTATTCAACGGCGCGAAG</b>
Foothold-3b	<b>TCTGAATGGTTAACAAACCGAAAGGAGCTGAAAAGTCGTGTATTAGA</b>
Foothold-3c	<b>TCTGAATGGTTAACAAACCGGAAGAAAGCGAAAGGTATTAAATGGTAGTTGATCCAATTAAGTT</b>
Foothold-2a	<b>TCTGAATGGGTACTGTGTAGCGAACCGACCGCCATTGAGCTTCGGAATCTGGCAAGTGTAGC</b>
Foothold-2b	<b>TCTGAATGGGTACTGTGTGTTATAACGGAAGTTGAGATTAAATATCGCG</b>
Foothold-2c	<b>TCTGAATGGGTACTGTGTGGTCACCGTCCAATAGTAAAAAGAAAAACCAAAACCCCTCGTTA</b>
Foothold-1a	<b>TCTGAATGGATTGATCAAACCTAAAACGAAAGAGGGCACAGGACGT</b>
Foothold-1b	<b>TCTGAATGGATTGATCAGAAGGCTTAAAATACGTATGCCCTATGG</b>
Foothold-1c	<b>TCTGAATGGATTGATCAGCTACAGGGCGCGTAACGCATAACCGGCCTCATCACC</b>
Foothold-6d	<b>TCTGAATGGTCAAGTGTTCATAGTTAGCGTAATCCACAGACAGGCCAGGTGTCCT</b>

Foothold-6e	<b>TCTGAATGGTCAAGTGTTGCTTATCACCGTACGCAATTGATC</b>
Foothold-6f	<b>TCTGAATGGTCAAGTGTTGCTTAGAATCAGAGCGTATAGCGTGCCGT</b> GTTTGCTCAAGAGATGAAA
Foothold-5d	<b>TCTGAATGGAAATCCTGTTAAGGTGAATTAAAGCGCAGTCTGAA</b> AAGCCAGAATCCACCAGATTAA
Foothold-5e	<b>TCTGAATGGAAATCCTGTTTAGACACCGGATTGCCATTGGTCAA</b> CTGTAGCAGAATCAAGTTT
Foothold-5f	<b>TCTGAATGGAAATCCTGTTAACGGTACGCCAGAGACTCCTTATATA</b> GCCAAATTATTCAATTAA
Foothold-4d	<b>TCTGAATGGAGTTGTTGATT</b> AATAACATAAAAACAGGGAATAAATA TTCTG <b>ATGCTTGATCTGGATCATAGCTACTGA</b> TTT <b>ACTTACTCACCTA</b> <b>AACACACATCA</b>
Foothold-4d.v2	<b>TCTGAATGGAGTTGTTGATT</b> AATAACATAAAAACAGGGAATAAATA TT <b>CGGTTCCAT</b>
Foothold-4e	<b>TCTGAATGGAGTTGTTGATT</b> ACAGAGAAAAATGAAAAGGTTTAG TAAAAGAGTCT
Foothold-4f	<b>TCTGAATGGAGTTGTTGATT</b> CCACCGGAAGCCTAAAGATTAACTGA G
Foothold-3d	<b>TCTGAATGGTTAACACCTT</b> TTATCTTAGCTAATGCAGTCTTACGTA
Foothold-3e	<b>TCTGAATGGTTAACACACT</b> TAACGTTAGTATAAAGGCCCTAAACAA TAGATAAG
Foothold-3f	<b>TCTGAATGGTTAACACCTT</b> GCAATACTTCTTGATGCGTTAAGAAA AAAATAAACGTGTGATAAAATT
Foothold-2d	<b>TCTGAATGGGTACTGTGTTAGTGA</b> ATTATCTGCTGAGAAGAGTC ATCAAGCC
Foothold-2e	<b>TCTGAATGGGTACTGTGTTGTTCTGAAAATTATTGCACTTAGAT</b> TA
Foothold-2f	<b>TCTGAATGGGTACTGTGTTGAGTAGAAGAACTCAATGATGATTCAA</b> AAAATCGCCTGATTGCATCG
Foothold-1d	<b>TCTGAATGGATTGATCATT</b> TAATGCGCATGGCAATTCAATCATATGA TTATCAGAAGAGCC
Foothold-1e	<b>TCTGAATGGATTGATCACAATATCAACTATCTAAAAAGTTGAAAAT</b> ATCTGGAACCTCAAATA
Foothold-1f	<b>TCTGAATGGATTGATCACGCCAGCCATTGCAACAGGGTGGACTCC</b>

**Supplementary Table S14.** DNA sequences of the locking strands for the large origami ring. The double-stranded stem region is colored red and the locking sequences are highlighted in orange. Toehold regions to initialize the toehold-mediated strand displacement reactions to open the locks are colored in purple. Foothold strand 4d (see Supplementary Table S13) serves as an additional lock.

Linker-3a	TCAGTAGCTATGATCCAGATCAAGCATTAGAACAGGTAGAAGGATA A
Linker-3b	TGATCCAGATCAAGCATTAGGGCGCTAGGGCGCGTACATAATATGTA TTGCTGAAGCAAACCTCCCTGATGCTTGATCTGGATCATAGCTACTGAT <b>TTTCATCCTCTACA ACTATCTACTC</b>
Linker-3c	AGAGGTCACCTTATTAGCTAAGCGCTGATGCTTGATCTGGATCATAG CTACTGATTTCATCCTCTACA ACTATCTACTC
Linker-1a	AGCTATGATCCAGATCAAGCATTAGGACGGATTGAGGGAGGGAAAGAT TAAATCCTGAGAAGTGCTGATGCTTGATCTGGATCATAGCTACTGATT <b>TACTTACTCACCTAACACACATCA</b>
Linker-1b	<b>TCAGTAGCTATGATCCAGATCAAGCATTAGTTTAAAAGAAAAACG</b> CACAAAGTTTAAGAAACAATAGCT
Linker-2a	TCAGTAGCTATGATCCAGATCAAGCATTAGGCTATTAGTC
Linker-2b	GAGGTGAAATCAACTATCTTAAAGATATGGAGACATCGCATG <b>CTGAT</b> <b>GCTTGATCTGGATCATAGCTACTGATTGACACTAACTCAAGTGTTCG</b> AGTA
Linker-2c	ACCGAACCTAAAAGCACTAATACCTGATGCTTGATCTGGATCATAGC TACTGATTGACACTAACTCAAGTGTTCGAGTA

**Supplementary Table S15.** Unlocking strands to open the locks of locking couples L1-L1', L2-L2' and L3-L3'. Toehold regions are colored in purple.

Open-L1	<b>TGATGTGTGTT</b> AGGTGAGTAAGT
Open-L2	<b>TACTCGAACACT</b> TGAGTTAGTGTC
Open-L3	<b>GAGTAGATAGT</b> TGAGAGGATGA

**Supplementary Table S16.** Foot strand for the surface modification of the AuNPs. The capture sequence to bind to the footholds is colored in green. The thiol-modification is shown in brackets.

Foot	CCATTCAGTTT[ThiC3]
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**Supplementary Table S17.** DNA sequences of the blocking and releasing strands. Blocking strands: green, foothold binding sequence; purple, toehold region. Releasing strands: purple, complementary toehold; green: sequence to deactivate blocking strands.

Blocking-1	TGATCGAATCCATTCCCTCACTTTCA
Blocking-2	ACACAGTACCCATTCCCTACCTATGT
Blocking-3	GGTTGTTAACCATTCATGTACTTAC
Blocking-4	TCAACAACTCCATTCAATGCCTGTGA
Blocking-5	ACAGGATTCCATTCCCATTATTAG
Blocking-6	AACACTTGACCATTCTAACTTCTTA
Releasing-1	TGAAAGTGAAGGAATGGATTGATCA
Releasing-2	ACATAGGTAGGAATGGGTACTGTGT
Releasing-3	GTAAGTACATTGAATGGTTAACACC
Releasing-4	TCACAGGCTATGAATGGAGTTGTTGA
Releasing-5	CTAATAATCGGAATGGAAATCCTGT
Releasing-6	TAAGAAGTTAGGAATGGTCAAGTGT

#### **IV. References**

1. Douglas, S. M.; Marblestone, A. H.; Teerapittayanon, S.; Vazquez, A.; Church, G. M.; Shih, W. M., Rapid Prototyping of 3D DNA-Origami Shapes with Cadnano. *Nucleic Acids Res.* **2009**, *37* (15), 5001-6.